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Research Article

THE BASICS OF BETAVOLTAIC NUCLEAR BATTERIES

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A B S T R A C T

From the extremely pollutant fossil fuels, to the problematic lithium batteries and the green wind turbines, new forms of generating energy are always necessary. To this day, new sources are being developed and improved to be applied in many different fields. Among those, betavoltaic nuclear battery (BNB) is a device that utilizes the radioactive decay to generate electricity. This work will present in a simple manner, the basics of BNB operation and necessary requirements to build one. Basic Radiation Physics Concepts, BNB configuration requirements, and a literature review were also given. At the end, a discussion containing the principal uses, examples on improving battery efficiency, and radiation damage is provided.

INTRODUCTION

Various sources of energy have been developed by mankind. From extremely polluting fossil fuels, to problematic lithium batteries, and green wind turbines. They contributed to the advancement of the human species, allowing to increase food production, expand civil construction, accelerate physical and virtual communication, and advance knowledge and science. New energy sources are being developed and improved for various applications. Among them, nuclear batteries are devices that use radioactive decay to generate electricity.

There are many types of nuclear batteries: thermoelectric(Ritz and Peterson, 2004), thermophotoelectric(Wang *et al*., 2020), direct charge(Jahangiri *et al*., 2021), thermionic(James *et al*., 2021), flicker-mediated(Prelas *et al*., 2016), direct energy conversion(Wang *et al*., 2019), alphavoltaic(Weaver *et al*., 2018), and betavoltaic(Uhm *et al*., 2016). This work will focus on betavoltaic batteries.

In 1913, Henry Moseley invented the first energy generator that used radioactive decay. It consisted of a silver glass sphere with a radio-226 source mounted in the center and an insulated electrode. Electrons resulting from the beta decay of radio-226 caused a potential difference generating an electric current. However, the current was too low for practical applications(Moscow Institute of Physics and Technology, 2018; Prelas *et al*., 2016).

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In 1953, Paul Rappaport proposed the use of semiconductor materials to convert beta decay energy into electricity. Beta particles emitted by a radioactive source ionize atoms in a semiconductor, creating decompensated charge carriers. In the presence of an electric field of a p-n structure, the charges flow in one direction, resulting in an electric current. Batteries powered by beta decay came to be known as betavoltaics(Moscow Institute of Physics and Technology, 2018; Prelas *et al*., 2016).

Radioactive isotopes used in nuclear batteries have a half-life ranging from tens to hundreds of years, resulting in almost constant power. Unfortunately, the power density of betavoltaic cells is significantly lower than that of their galvanic equivalents. Despite this, betavoltaic began to be used in the 1970s in electronic circuits that demand little load (Moscow Institute of Physics and Technology, 2018; Prelas *et al*., 2016).

Interest in the use of betavoltaic batteries has grown substantially in recent years due to the prospects for use in microelectromechanical systems (MEMS). New generation MEMS and semiconductor devices require miniature power supplies and long operating times(Bykov *et al*., 2017).Often these devices are installed in places where maintenance is difficult, such as space operations, seabed exploration, and oil removal at the pre-salt layer.

Today several prototypes of betavoltaic microbattery based on different semiconductor materials such as silicon(Lei *et al*., 2014), silicon carbide(Coutinho *et al*., 2021), gallium nitride(Lu *et al*., 2011), and diamond(Bormashov *et al*., 2018) have been reported in the literature. Semiconductors with wide gaps are preferable for manufacturing power converters, because the conversion efficiency, η_{conv}, increases(Fradkin *et al*., 1965).Devices based on these materials can operate at elevated temperatures and are generally more resistant to radiation. This makes the operation possible in adverse conditions.

These prototypes described in the scientific papers mostly contain summary descriptions due to the strong presence of competition in the areas of application and industrial secrecy. Prelas*et. al*. (Prelas *et al*., 2014)stated in their review "The difficulty in following alpha and betavoltaic cell research literature is that most studies fail to provide enough information for a reader to fully understand the experiment and to properly interpret the results".

This work has the goal to present, the basics of BNB operation and necessary requirements to build one. Basic Radiation Physics Concepts, BNB configuration requirements and a literature review are given next. At the end, a discussion containing the principal uses, examples on improving battery efficiency, and radiation damage is provided.

Basics of betavoltaic nuclear batteries

Types of Nuclear Batteries

The known ways to directly convert nuclear energy into electrical energy can be classified into three major groups: radioisotopic thermoelectric generator (RTG), betavoltaic or alphavoltaic batteries, and photovoltaic devices(Keller, 1989; Magill and Galy, 2005; Prelas *et al*., 2016; Prelas *et al*., 2014; Wang, 1969).

RTG consists of converting the thermal energy from radioactive decay into electrical energy using the Seebeck effect. The disadvantages are the low general conversion efficiency (typically between 3-8%) and the need to use a radioactive source with very high activity (typically in the order of a few hundred kCi/TBq, unit of radioactive activity). However, it is possible to obtain electrical powers in the range between tens and hundreds of watts(Keller, 1989; Magill and Galy, 2005; Prelas *et al*., 2016; Prelas *et al*., 2014; Wang, 1969).

On the other hand, the other modalities consist of obtaining electrical energy from the interaction of a material medium with beta or alpha particles or with photons generated by secondary interactions(Corliss and Harvey, 1964; Daruich de Souza, 2020; Vértes *et al*., 2011; Wang, 1969).Although the output power of the betavoltaic battery is relatively low, the use of some type of electric accumulator (such as a chemical cell or capacitor) or the stacking of several modules can increase the output power from milliWatts to Watts(Bao *et al*., 2012; Bormashov *et al*., 2018).

Betavoltaic batteries should not be confused with radioisotopic thermoelectric generators. RTG cells convert the heat released by radioactive decay into electricity using thermocouples. The efficiency of RTGs depends on temperature and the output power is much higher than that of betavoltaics.

Nuclear batteries can operate continuously without recharging or replacement for years or even decades (depending on the radioisotope's half-life) and under extreme high temperatures. They offer exclusive advantages for use under these conditions(Bormashov *et al*., 2018; Corliss and Harvey, 1964; Magill and Galy, 2005; Moscow Institute of Physics and Technology, 2018; Prelas *et al*., 2016).

Efficiency

Any nuclear battery consists of a radioisotope source and a converter that is characterized by its total efficiency (η) that can be separated into two parts:

$\eta = \eta_{source} . \eta_{conv.ef.}$

η_{source} is the total power entering the converter divided by the total power produced at the source by the radioisotope and $\eta_{conv.ef.}$ is the total electrical energy output of the battery divided by the total power entering the converter (conversion efficiency)(Bormashov *et al*., 2018).

The most important goal of nuclear battery systems is to achieve greater efficiency with a smaller size. Performance is determined by the characteristics of the radioisotope, radiation transport, energy conversion transducers, electric coupling, and charge collection efficiency. The specific energy density (J/kg) of radioisotopes is inherently higher than chemical energy by many orders of magnitude, due to nuclear decay energy. However, according to the type of radioactive decay (alpha, beta, fission, etc.) the energy density varies inversely with the isotope half-life- i.e., the shorter the half-life, the greater the power density (W/kg). This fundamental principle makes the two desired properties of a nuclear battery, long shelf life and high power density, opposite(Prelas *et al*., 2014). Another important consideration is that the size of the device depends on the range of a particular particle in a specific material, called the linear range of radiation transport ($\lambda_{Rad.Trans.}$),and the volume of energy conversion in the transducer, referred to as the transducer scale length (λ_{Trans}).Both should be approximately equal, because this guarantees that a larger amount of particles are collected. This fundamental property is the main factor that determines the efficiency of a nuclear battery: systems that meet this requirement are more efficient, while systems that don't are less efficient. Achieving the "well-matched" scale of lengths is one of the main challenges encountered in trying to miniaturize systems(Prelas *et al*., 2014).

The variables that influence λ_{Trans} include: mass, charge, angular distribution, energy distribution of the source particles, atomic number, density, ionization potential of the material target, and the mechanisms by which the particle interacts with the target. These collectively cause $\lambda_{Rad.Trans.}$ to vary widely between radioisotopes. The factors that determine λ_{Trans} include the battery's energy conversion mechanism, the mechanical and electrical properties of the target material, and the effect of radiation damage on the target(Prelas *et al*., 2014).

Choice of isotope

The decision of which isotope to use in a nuclear battery is complex. First, the type of radiation that the isotope emits will determine whether or not there is a good match between the range of the emission and the length of the transducer. Second, the half-life of the isotope will determine the initial necessary radioactive activity of the source, as well as the effective useful life of the nuclear battery, as long as there is no damage to the materials caused by the radiation. The third consideration is the radiation decay energy, which together with the activity determines the effective power. The fourth consideration is how the isotope is produced (figure 1), which determines its cost. If the isotope is produced naturally as part of the decay chain of uranium-238, uranium-235, or thorium-232, it is easily found (although high values can be spent on purification/concnetration). If the isotope is a by-product of fission, then it can be recovered from the nuclear fuel that has already been used. If the isotope needs to be produced in a nuclear reactor or in accelerators, its cost can be high (due to the high cost of enriched targets and purification routes). The fifth consideration is whether or not other forms of radiation are being emitted by the isotope (for example, gamma rays). Gamma rays are highly penetrating and therefore require shielding to protect people, the environment, and electronic devices. Shielding can compromise the portability of the equipment(Daruich de Souza *et al*., 2021b; Prelas *et al*., 2014).

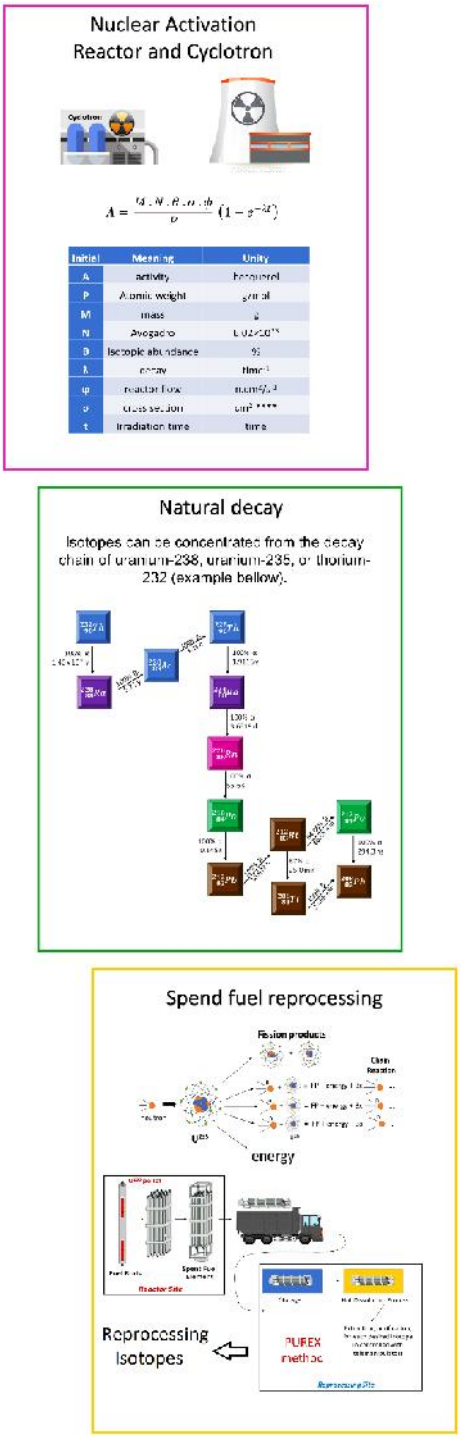


Figure 1 Isotopes Production routes.

Beta emission

Beta emission, beta disintegration, or beta decay is the process by which an unstable nucleus can be transformed into another nucleus by emitting a beta particle, changing its number of protons and number of neutrons. Beta emitters are located at the bottom of the line of stable elements in the periodic table of radioactive elements (figure 2). (KAPLAN, 1978)

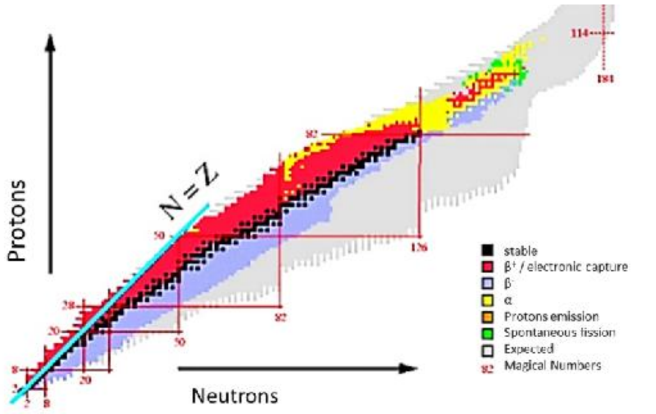


Figure 2 Table of radioactive elements. The black line is the stable isotopes and the lilac portion, the beta emitters. Adapted from (LEIFIphysik and Stiftung, 2021).

In β^{-} decay, a neutron is converted into a proton, with the emission of an electron and an antineutrino (the antiparticle of the neutrino). As a consequence, the atom is changed:

$$n \rightarrow p + e^{-1} + \bar{\nu}$$

$${}^{A}_{Z}X \rightarrow {}_{Z+1}^{A}X' + e^{-1} + \bar{\nu}$$

Neither the beta particle nor its associated antineutrino exists within the nucleus before beta decay, but are created in the process of decay maintaining the energy conservation principle. By this process, unstable atoms obtain a more stable relationship between protons and neutrons. The probability of emission is determined by its nuclear bonded energy. Beta decay is a consequence of the weak force, and is characterized by relatively long decay times (KAPLAN, 1978).

Compared to heavy ions, the path of an electron in matter is complex due to Coulombian interactions. As the incident electron has a mass equal to that of the electrons in the target, the electron is significantly scattered and follows the random path shown in figure 3 (KAPLAN, 1978).

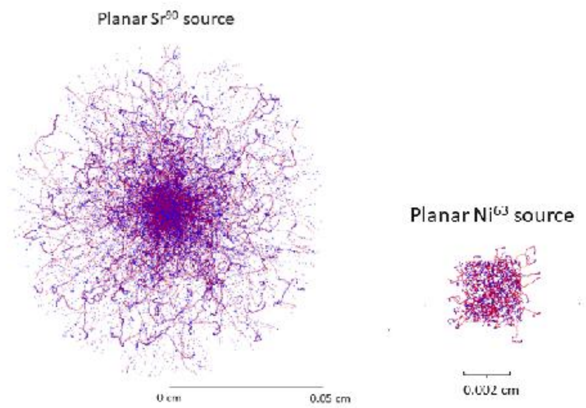


Figure 3 Plot of MCNP simulation of β^{-} decay from strontium-90and nickel-63 in a planar source geometry.

The ion pair that is produced from the interaction of beta particles with matter includes a secondary electron that is ejected from the atom's orbit. Secondary electrons normally have energies in the keV range (KAPLAN, 1978). Tertiary, quaternary or higher electrons can also be generated. Secondary gamma or X rays can demand increased shielding.

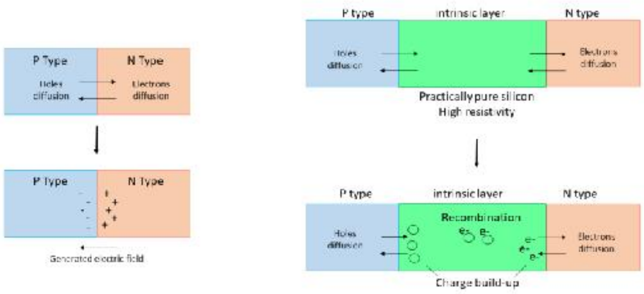
Accurate range calculations are essential when designing the battery in order to match the active region of the transducer (L_{Trans}) in the ideal position to harvest the energy of the beta particle ($\lambda_{Rad.Trans.}$). To calculate the range of a beta particle in matter, the full spectrum of beta energy must be used in the model (formulas using approximations should not be used). Mostly, Monte-Carlo Simulation is used to study this matter (Prelas *et al.*, 2014). Monte-Carlo Simulation is a powerful tool to predict how the device will behave, in terms how many particles will hit the transducer, how much will contribute to power generation, the heat and current generated, and the necessary shielding for the device. Different codes such as mcnp, GEANT4, Penelope, EGSnrc, Fluka and others are available.

The great difficulty with betavoltaic batteries is that the technique is still in the development stage and/or the description in the literature is scarce and incomplete(Prelas *et al.*, 2014).Most studies do not provide enough information for experiments, data and results to be interpreted. For example, information such as the type of radioactive source used (pure or in a mixture format), how the source is coupled to the cell, the dimensions of the source, which type of junction p-n, are often not presented (Prelas *et al.*, 2014).In other cases, a vague description can be found in patents mostly due to industrial secrecy.

The electrical powers generated by these devices make up a wide range of values, ranging from nano to milliWatts depending on the design of the device, but which, in all cases, are limited to low electrical powers(Keller, 1989; Prelas *et al.*, 2016).To circumvent this issue and increase the power, hundreds or thousands of cells / modules can be built.

Semiconductor diodes

In addition to the beta emitting radioisotope, betavoltaic cells are composed of semiconductors with a p-n, p-i-n junction, as shown in figure 4, or Schottky type diode.



a) **b)**

Figure 4 a) p-n junction. Note the formation of the potential barrier after the diffusion of the charges. b) P-i-N Diode. Part of the charge recombines and part is accumulated, decreasing the resistivity of region I.

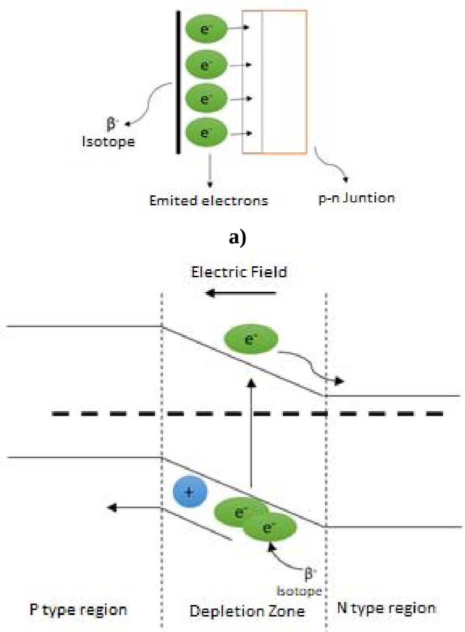
p-n diodes

Crystalline materials have three energy bands referring to their conductivity. The first is called n-zone, valence band, or donor zone and it has a lattice containing the silicon atoms (for example) with an impurity, such as phosphorous. This impurity contains and extra electron in the valence layer. This electron is free to move around in the lattice. The second in called p-zone, conduction band, or accepting zone and it has a lattice containing the silicon atoms with an impurity, such as boron. This impurity is missing an electron in the valence layer. This hole is free to move around in the lattice. When this bands are in contact, electrons form the n-side can jump to the p-side(AHMED, 2007).

The third band is called depletion zone, p-n junction,forbidden zone, or bandgap. Once the initial electrons jump to filled the initial holes, the potential difference of the area increases, i.e., is more difficult for the latter electrons to jump and fill further holes. In this region there are no charge carriers (electrons and holes). Insulating materials have a large width prohibited zone between de valence-conduction bands, preventing the electrons jumps, therefore they cannot conduct electricity. The conductive materials have an almost null prohibited zone, which facilitates the penetration of electrons from the valence region into the conduction region, therefore, easily conducting electricity. The semiconductor material, on the other hand, has anintermediate forbidden zone and under special conditions can also conduct electricity(The National Programme on Technology Enhanced Learning (NPTEL) *et al.*, 2019; Vavilov and Ukhin, 1977). The effect caused by an electron emitting source is shown in figure 5.

By adjusting the density of impurities present in p-type and n-type materials, the width of the depletion zone changes. Normally, the width of the depletion in a well-designed cell will be about 1 mm thick. The challenge is to deposit the maximum energy of the beta particles from the source in the 1 mm depletion layer (Prelas *et al.*, 2014).

When radiation strikes, ionization occurs in the depletion region. Because there is an electric field inside, the new electron-hole pairs are attracted by the p and n regions, creating an electric current (Ji and Kim, 2021; Nam *et al.*, 2006; Neudeck, 1983).



b)

Figure5 Betavoltaic effect a) Schematic diagram of the betavoltaic effect b) Potential diagram for the betavoltaic effect. Adaptedfrom(Hang and Lal, 2003).

p-i-n diodes

P-i-n type diodes have an intrinsic layer between p and n layers. This layer is purely silicone. The p and nlayers are highly doped because they are used as ohmic contacts. Part of the charges recombine when they enter the intrinsic region. Part is accumulated, decreasing the resistivity of region i.In a p-i-n diode the depletion region exists almost completely within the intrinsic region. This depletion region is much larger than in a p-n diode and almost constant-size, independent of the reverse bias applied to the diode. This increases the volume where electron-hole pairs can be generated by an incident charged particle. So, the p-i-n diode provides additional sensitivity and performance over that of the basic p-n junction photodiode (Agarwal and Lang, 2005). The wider depletion width also improves the collection efficiency.

Schottky diodes

In Schottky type diode, when a metal or superconductor makes close contact with a semiconductor, the Fermi levels in the two materials must be equal in thermal equilibrium. The vacuum level must also be continuous. These two requirements determine a unique energy band diagram for the contact, as shown in Figure 6. The resulting band undergoes a curve at the interface creating a potential barrier known as the Schottky barrier. The height of the band, ΦB, is the difference between the metal working function, ΦM, (the difference in energy between the Fermi level and the vacuum level) and the semiconductor electronic affinity, X, (the difference between the edge of the semiconductor conduction band and the vacuum level). The width of the Schottky band depends, among other things, on the doping density of the semiconductor. Therefore, it is possible to adjust the barrier to allow more or less electrons to pass through.(Warwick Department of Physics, 2019)

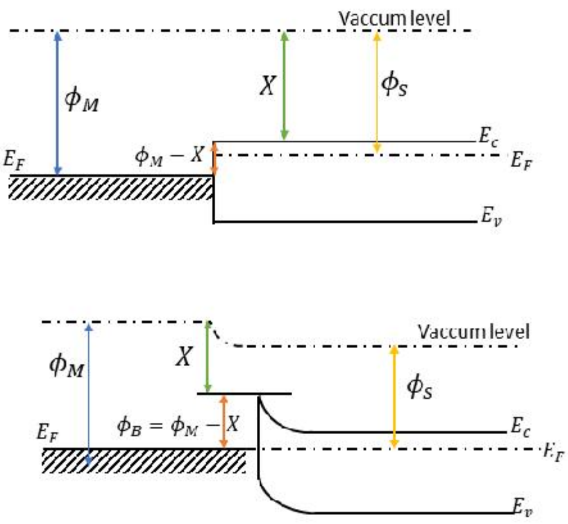


Figure 6 (a) Energy band diagram of a metal and a type n semiconductor under non-equilibrium conditions. (b) energy band diagram of a thermally balanced metallic semiconductor contact. Adaptedfrom(Warwick Department of Physics, 2019).

Output power

Once an electric field is established, electric current is generated in the system. The absolute efficiency of a device that converts energy is defined as:

$$\eta_{ab} = \frac{P_{out}}{P_{total}}$$

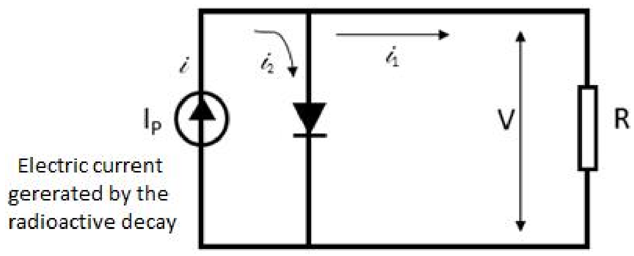
$$P_{out} = V \,.\, I$$

$$P_{total}(W) = A \,.\, E_{dis} \,.\, 1{,}6 \,.\, 10^{-19}$$

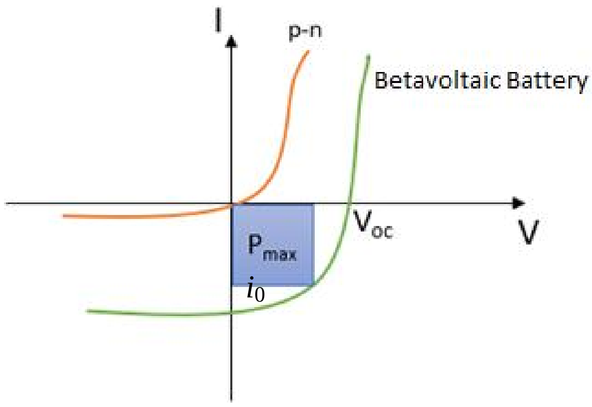
where P_{out} is the output power, P_{total} is the total power deposited in the energy conversion regions, V is the cell potential (volts), I is the cell current (amps), A is the radioactive activity (Becquerel, decays per second), E_{dis} is the energy produced by the decay (eV / Bq) and η_{ab} is the absolute efficiency. (Prelas *et al*., 2014)

To estimate current I, the maximum current created in the cell must be found. The maximum current is dependent on the energy transport efficiency to the depletion zone. This can be expressed by η_d which represents the fraction of power that is deposited in the depletion zone. Monte-Carlo-based transport codes are used in the calculation(Prelas *et al*., 2014).A simple electrical circuit model is shown in figure 7.

$$P_{absorbedinthedepletionlayer}(W) = P_{total} \cdot \eta_d$$



a)



b)

Figure 7 a) Circuit model; b) Voltage (V) x Current (I) curve showing the values to achieve P_{max}.Adapted from(Hang and Lal, 2003).

A small junction leakage current p-n (I_0) is required to achieve higher V values. For photovoltaic cells the value of I is usually in the range of 1-100 mA. Therefore, the leakage current is not important and could be in the order of nA or even µA. However, for betavoltaics with activities in the range of 1-100 mCi (37-3700 MBq), I_p is in the order of pA or nA. If the leakage current I_0, is in the range of nA or µA, there will be minimal output power.

Examples of Betavoltaic Batteries

The most used beta emitting radioisotopes are*:

$$Ni^{63} \rightarrow Cu^{63} + \beta^{-} E_{med} = 17.4\ keV; T_{\frac{1}{2}} = \ 101.2\ years$$

$$H^{3} \rightarrow He^{3} + \beta^{-} E_{med} = 5.7\ keV; T_{\frac{1}{2}} = \ 12.3\ years$$

$$Pm^{147} \rightarrow Sm^{147} + \beta^{-} E_{med\beta} = 61.9\ keV; T_{\frac{1}{2}} = \ 2.6\ years$$

$$Sr^{90} \rightarrow Y^{90} + \beta^{-} \rightarrow Zr^{90} + \beta^{-} E_{medSr} = 195.8\ keV; T_{\frac{1}{2}} = 28.79\ years\ **$$

* for more information, including older models with other radioisotopes, see reference (Bykov *et al*., 2017).
** due to the Sr^{90}'s high power density, it is used more in RTG models than BNBs.

Tritium models

Andreev *et. al*. (Andreev *et al*., 2000)presented a model (figure 8) using tritium. An AlGaAs semiconductor was used with an ultra-fine depletion zone (0.01 -0.03 pm). The results reported were:

- output current was 0.75-1 µA/cm^2;
- the circuit voltage was 0.65-0.93 V;
- the maximum output power was 0.55 µW/cm^2;
- the maximum specific output power reached was 335 nW/Ci (9.05 nW/GBq);
- no detailed information about the source.

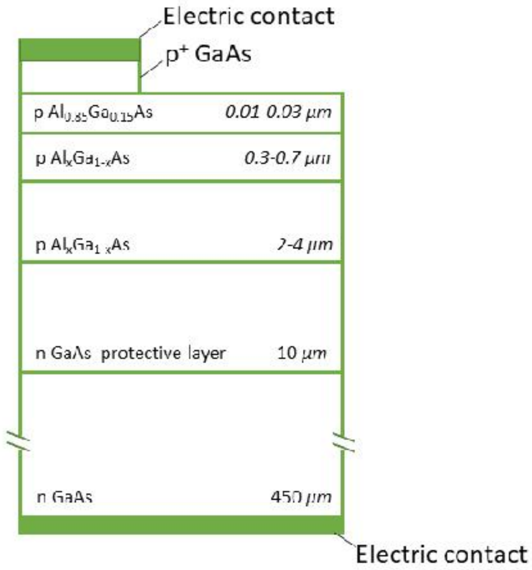


Figure8 Andreev *et. al*. betavoltaic battery.

Clarkson *et. al*. (Clarkson *et al*., 2007)used tritium in gaseous state in their battery model. It was reported:

- p-n junction of macro porous silica: pores with 1.1 µm in diameter and 2.5 µm spacing;
- Estimated efficiency was 0.22 %.
- Deus (Deus, 2000)reported ultra-thin tritium batteries:
- 5 mm p-n junction;
- radioactive tritium gas was used;
- circuit current was 637 nA/cm;
- Circuit voltage of 457 mV;
- Maximum output voltage 136 nW/cm^2;
- Several problems with semiconductor failures caused by radiation have been found.

Nickel-63 models

Accordingly with Bykov*et al*. (Bykov *et al*., 2017) strontium-90 and promethium-147 have β- emission energies that can affect the radiation resistance of most substances. Other possible isotopes, such as Carbon-14, has a long half-life (5700 years) and therefore the electron flow is of low intensity. Since tritium is a gas, using it to create a high-density power supply is highly complex. Thus, the nickel-63 isotope is the most suitable material, with the best prospects for use.

Lu *et. al*. (Lu *et al*., 2011)presented a model using nickel-63 and a gallium nitride (GaN) transducer. The scheme is shown in Figure 9. The parameters obtained were:

- GaN films were manufactured on sapphire substrates by metal-organic chemical vapor deposition;
- Voltage was 0.1 V
- Current density was0.35 ~1.2 nA/cm^2;
- The conversion efficiency achieved was 0.32 %;
- The activity used was 33 µCi/mm^2(1.22 MBq/mm^2) ;
- Current was 4 pA.

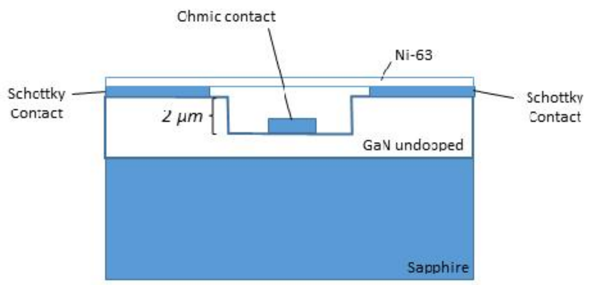


Figure9 Lu *et. al* betavoltaic battery scheme.

The design, manufacture and testing of a betavoltaic battery with Schottky transducers was done by Li *et. al*. (Li *et al*., 2011) (figure10). Nickel-63 was 4 mCi/cm^2(148 MBq/cm^2) was used in the project, which had a total area of 3.14 mm^2.

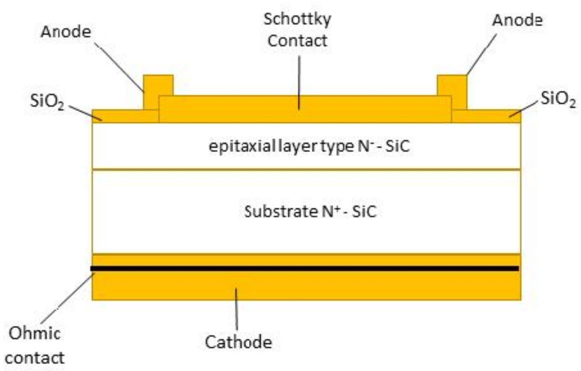


Figure10 Li *et. al* betavoltaic battery scheme.

The characteristics presented were:

- The circuit voltage was 0.27 V;
- Current density was 25.57 nA/cm^2;
- The maximum power was 4.08 nW/cm^2;
- Power conversion efficiency was 1.01 %.

Zaijun*et. al*.(Cheng *et al*., 2010)presented a nickel-63 betavoltaic microbattery with a GaN semiconductor with a wide depletion band. The scheme is shown in figure 11.

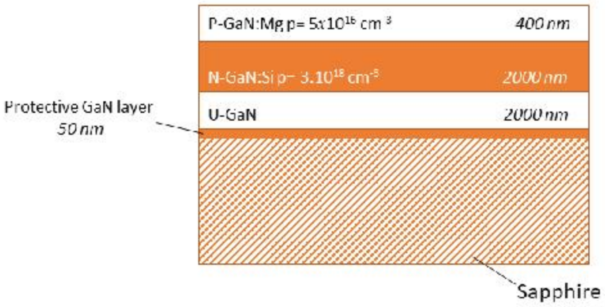


Figure 11 Schematic of the GaN p-n junction. The development of the device was the result of several doctoral and postdoctoral work.

Cells were fabricated using metal-organic chemical-vapor deposition. Organic chemical vapor deposition of metals (MOCVD) is a well-known and controllable method of synthesis. The reaction is carried out with the phases in vapor form, which allows the control of the ratio of the regents, growth time and growth rate. The layers are deposited by transporting different precursors or reactants in the vapor phase, under controlled pressure, to a reactor chamber (vertical, horizontal, or barrel) (Donkor, 2001; Zhang *et al*., 2016).Results were:

- 11 mCi (407 MBq) of nickel-63;
- Open circuit voltage - 25 mV;
- Current2 nA;
- Cell size 1x1 cm;
- The author presents the following reasons that justify the unsatisfactory result: low bandwidth obtained (should be in the order of µm), the 1x1 cm size is too large, and increasing defects in the depletion zone resulting in poor quality of the GaN crystal;
- Power results were not presented.

Hang *et. al*. (Hang and Lal, 2003)present theoretical and experimental studies on betavoltaic microbatteries using nickel-63 thin films with 100 mCi (3.7 GBq) of activity. Output power obtained is in the order of nanoWatts. Two types of p-n junction were tested: silicon p-n diode with nickel-63 electrodeposited and micromachined p-n junction structure in bulk with inverted pyramid arrangement. The results were:

- Power obtained is on the scale of one nanoWatt;
- 0.25 mCi (9.25 MBq) of nickel-67;
- 128 milliVolts open circuit voltage;
- 2.86 nA short-circuit current;
- According to the author, the output power can be increased using greater radioactive activity and efficient collector designs.

Duggirala*et. al*. (Duggirala *et al*., 2008)demonstrated a betavoltaic generator with an energy conversion efficiency of 5.1%. Silicon converters with reciprocal piezoelectric elements were used. The results were:

- Power generated by the radioisotope: 0.94 µW;
- 9 mCi (333 MBq) of nickel-63;
- Electrical output power: 22 nW;
- Piezoelectric output power: 750 µW;
- The authors claim that their prototype can last 100 years and be used in wireless microsensors;
- The author claims that he overcame the low power output with the use of electromechanical generators with pulsed power output (although the publication does not offer further details or reference);

Qjao*et. al*. (Qiao *et al*., 2011)used a Schottky-type diode (SiC) in the construction of their microbattery. The model is shown in Figure 12.

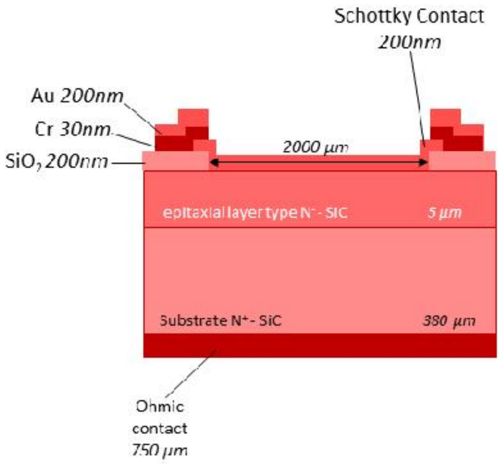


Figure12 Schottky diode cross section.

The results obtained were:

- Current density in the circuit: 13.82 nA/cm^2;
- Open circuit voltage: 0.26 V;
- Output current density: 10.76 nA/cm^2;
- Output voltage: 0.19 V;
- Output power: 2.04 nW/cm^2;
- Efficiency: 0.5 %.

Bormashov*et. al*. (Bormashov *et al*., 2018)presented a prototype betavoltaic battery consisting of a nickel-67 sheet and 200 vertically stacked single-conversion cells based on diamond diodes. When the long half-life of nickel-63 is considered, the specific energy is approximately 3300 mWh/g (an order of magnitude greater than commercial chemical batteries). The authors were able to fabricate diamond converters less than 100 µm thick using plasma and mechanical polishing (more design references (Delfaure *et al*., 2016; Tchouaso *et al*., 2018; Trucchi *et al*., 2008)). Model is presented in figure 13.

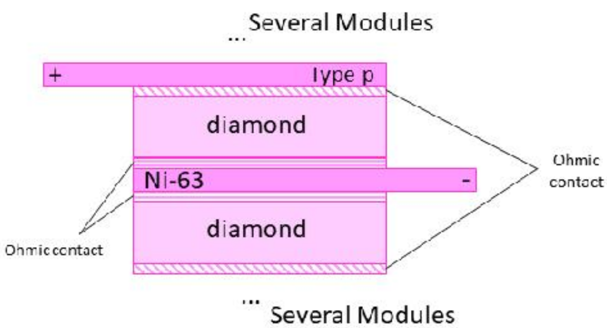


Figure 13 Schematic view of the prototype.

The results obtained were:

- The maximum electrical output power of 0.93 µW was obtained in a total volume of 5x5x3.5 mm^3;

- Output power density:10 μW/cm^3;
- Cells connected in parallel generated 1μA;
- A 200 nm gold layer was deposited on both sides of the connectors to ensure better electrical contact;

Our research group develops a nickel-63 battery model that is modular. The picture of the device can be seen in figure 14.

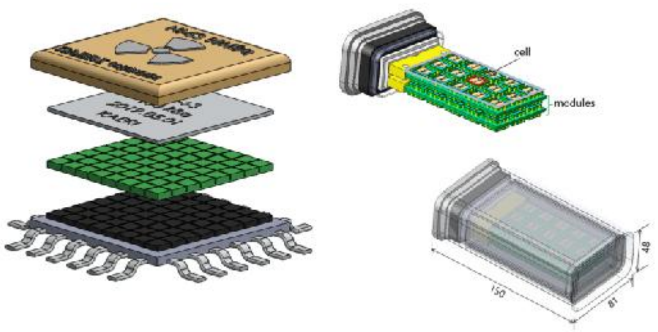


Figure14 Betavoltaic battery models being developed in South Korea.

The manufacturing process begins with obtaining the precursor, Niquel-62. The material is subjected to the Neutron flux of the Hanaro Reactor (30 MeV, Kaeri - Daejeon, South Korea) activating it in nickel-63. Each battery cell contains nickel-63 which has been electrodeposited on a sheet of natural non-radioactive nickel (nickel-62). The electrolytic bath is carried out in the presence of boric acid, sodium chloride and saccharin. The complete process takes about a day and the thickness achieved is 2 μm. The manufacturing process for P-i-N type diodes is carried out in partnership with ETRI - Daejeon, South Korea.

The voltage x current curve obtained is shown in figure 15.

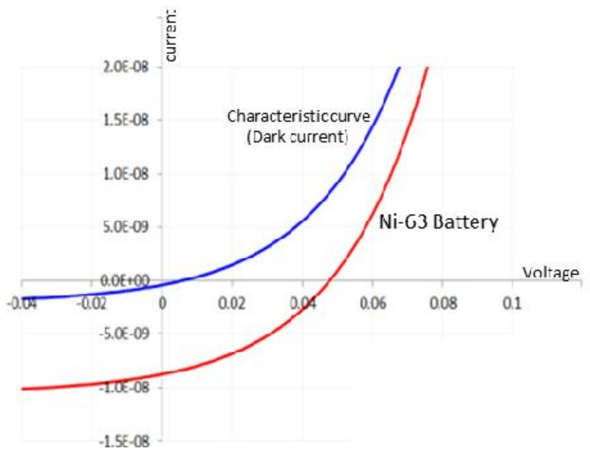


Figure15 voltage vs. current curve for the batterybeing developed in South Korea.

The results obtained for a cell:

- Open circuit voltage: 69.2 mV;
- Short circuit current: 9.74 nA;
- Output current density: 60.9 nA / cm^2;
- Output power density: 4.21 nW / cm^2;
- Output power: 0.67 nW.

Promethium-147 models

Flicker *et. al.*(Flicker *et al*., 1964)featured a battery model using promethium-147. The author highlights the project's difficulties, among them, the difficulty of synthesis (decomposition with the use of urea), manipulation, purification and preparation of the radioactive layer. The efficiency achieved was 0.7% with the activity of 6.8 Ci (251.6 GBq) in an area of 1 cm^2.

Kavetskiy*et. al.* (Kavetskiy *et al*., 2011)presented a direct charge betavoltaic battery. The source of charged particles directly connected to a charge collector through a dielectric element (capacitor). This system manages to accumulate high efficiencies (≈15 %) generating high voltage. The promethium-147 activity used ranged from 1.5-9.6$x10^{10}$ Bq. The study, which is in its initial phase, reached the following data:

- For parallel plate capacitor: 18 kV (3.5% efficiency);
- For steel cylinder capacitor: 28 kV (8% efficiency);
- For aluminum cylinder capacitor: 45 kV power (15% efficiency).

4^{90}Sr/^{90}Y models

The possibility of using the beta decay of the ^{90}Sr/^{90}Y father-daughter isotope pair started to be investigated recently. Advantageously, both decay predominantly by beta radiation. Yttrium-90 has an emission of 933.7 keV (99.98%) energy, contributing to the generation of charges in the transducer. The difference in theoretical efficiency is shown in Figure 16. The major issues are radiation damage provoked by the high bremsstrahlung emission and the heat damage triggered due to the high heath released during decay. For this reason, ^{90}Sr/^{90}Y is more suitable for use in RTG devices.

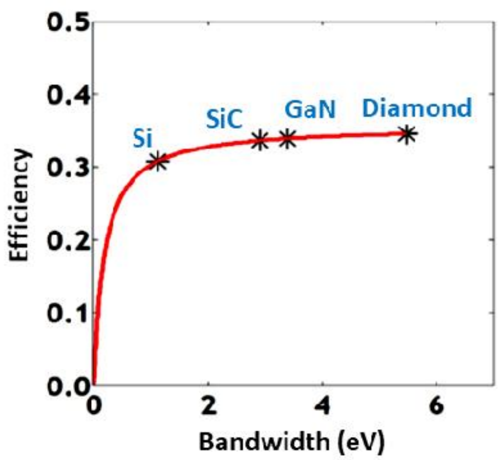


Figure16 Difference in theoretical efficiency for different semiconductors for ^{90}Sr/^{90}Y batteries.

Dixon *et. al.* (Dixon *et al*., 2016)evaluated the potential of a ^{90}Sr/^{90}Y battery using a diamond semiconductor. The device would have 1 W of strontium-90 power with a conversion efficiency greater than 10%. The authors devised a model that modularly stacks the device with alternating 45 µm-thick strontium-90 layers with 265 µm-thick p-n diodes (figure 17). For a device with an area of 25 cm^2, 35 modules would be needed to generate 1 W at 10% efficiency. Individual devices can then be connected in series or in parallel. A collection of such devices can generate 100 W would weigh less than 5 kg.

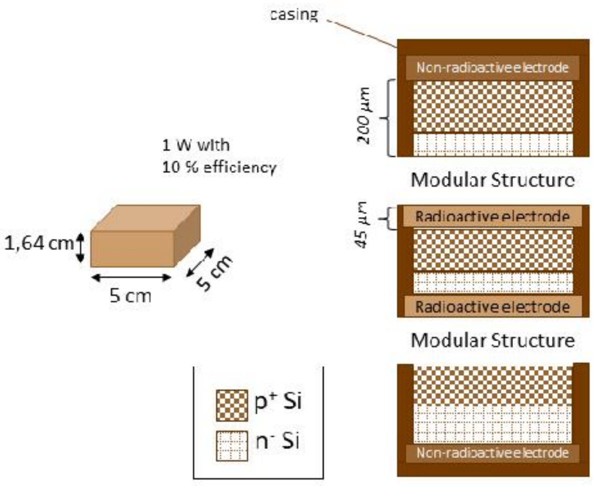


Figure 17 General dimensions and cross section of the proposed 1 W device.

DISCUSSION

Radiation Damage

The interaction of radiation with matter causes damage to materials. This damage can be permanent or temporary and results in a wide variety of chemical and physical effects. Depending on the quantity, type of radiation and energy, the effects of radiation can, for example, degrade the performance of devices, kill cells or weaken materials (Daruich de Souza *et al.*, 2021a).

It is extremely important to study these effects. Predicting the rate of degradation of materials is directly linked to the durability and efficiency of the nuclear battery device (Daruich de Souza *et al.*, 2021a). There are several forms of interaction between radiation and matter. But basically, it can:

- To suffer a change of direction due to electric fields (Rayleigh and Thomson scattering);
- Interact with the electronsphere (photoelectric effect, Compton effect and production of pairs / triplets);
- Interact with the nucleus of the atom (nuclear reactions);
- Interact with the atom as a whole (Knoll, 2000; Pessoa *et al.*, 1978).

Non-ionizing energy loss (NIEL) processes are interactions in which the energy transferred by the incident radiation does not result in ionization, but has an effect on the crystalline network of the material. Direct collisions may be sufficient to remove an atom from the crystalline lattice or create a transfer not strong enough to move the atom from its location in the lattice, dissipating energy in aslattice vibrations (phonons) (Daruich de Souza *et al.*, 2021a; Leroy and Rancoita, 2007).

In low radioactive environments, radiation damage by NIEL can diminish, for example, the thermoelectric conversion efficiency in nuclear batteries. In Wang *et. al.* (Wang and Leonard, 2021) commercially available bismuth telluride materials showed significant changes in individual properties. N-type materials electrical properties and p-type materials thermal conductivity were affected by irradiation-induced defects. This might result in radiation levels restriction, affecting device usability. In betavoltaic devices, protection layer of the p-i-n diode is possible, but the device will have a lower output power due to shielding of the desirable beta particles. Beta particles are exponentially attenuated for most of their reach in a material medium, and the attenuation coefficient has a dependence with the maximum energy of the beta spectrum.

Improving efficiency

This is a topic with vast discussion in the literature with subjects ranging from silicon diode design, electric coupling adjustments, metallic contacts improvement, radiation damage prevention, radiation source fabrication techniques, deposition studies, among others.

The matter also depends on the manufacture plans of the developers. For example, in application as sensors in space, space radiation will require appr ate shielding, impacting the final size of the sensor. This might bring issues on the type of diode used. Another example, for use in cryptography (by generating random numbers) signal noise is a major aspect of the project. The electronics used might directly affect the amount of radiation material used and type of diode.

Next, 2 examples of works that improved the betavoltaic battery electrical performance are summarized.

Ulmen at. al. examined the operation principle of a p-n junction nuclear battery and means to improve its performance (Ulmen *et al.*, 2009). The performance characteristic of a 4 mCi (148 MBq) Nickel-63nuclear battery was tested using a picoammeter/voltage source. The open circuit voltage was found to be 0.8 mV while the short circuit current was 11 nA. The maximum power output was found to be 2.5 picowatts at a voltage of 0.4 mV. While this performance is relatively low, the authors claimed that it demonstrates the validity of the fabrication technique of the battery. The junction depth and doping densities of the n and p regions were tested to improve power. The epitaxial film growth technique was used and yielded a smoother surface, helping with coupling. Metallic contacts manufacture was improved by fabricating on both sides of the wafer that serve as ohmic and seed layer. Standard photolithography and lift-off technique were used to form both contacts. The ohmic contacts were formed on the undoped p-type side of the wafer. The fabrication route was: 200 Å of sputtered Aluminum, annealed at 450 ºC for 30 min under N_2 atmosphere, with a 100 Å Chromium layer and 1,500 Å Gold layer placed over it. On the reverse side, the side of the p–n junction, photolithography and lift-off techniques were again used to form the Nickel-63deposition seed layers using 500 Å of sputtered Titanium and 200 Å of sputtered Nickel (Ulmen *et al.*, 2009). The authors say that adjusting the P–N junction and depletion regions depths are keys to higher device performance (Ulmen *et al.*, 2009).

Xue*et. al.* (Xue *et al.*, 2019)proposed and experimentally validated different techniques to improve nuclear batteries power conversion efficiencies (PCE). A thin scintillator layer has been added into the battery structure to boost PCE through the combination of indirect (light generation) and direct (electron–hole pair generation) collection of radiation energy. The scintillator layer also serves as an energy degrader to reduce the charged particles energy, consequently increasing their stopping power and energy deposition in the sensitive region of the devices. The phosphor layer added between the radioisotope source and the semiconductor device resulting in 4 times more power in their new 3D device that uses SiC. The maximum output power density was 1.19 nW.cm^{-2} with a source activity of 0.4 µCi (14.8 kBq).

Application

Interest in the use of betavoltaic batteries has grown substantially in recent years due to the prospects for use in microelectromechanical systems (MEMS). New generation MEMS and semiconductor devices require miniature power supplies and long operating times(Bykov *et al*., 2017).

In the 1960's, the idea of bringing nuclear batteries into the pacing industry was first introduced and ultimately pursued in the 1970's. Several pacemaker manufacturers had introduced nuclear models. They provided young patients the opportunity to have one pacemaker last their entire life, without surgery to change the device battery. These nuclear pacemakers also proved cost-effective in comparison to the lithium battery powered pacemakers of today as follow-up costs of the two are roughly $19,000 versus $55,000 respectively (Emery, 2007). Ultimately, the implementation of nuclear pacemakers came to a halt in the mid 1980's as lithium-powered pacemakers containing new technology and a more feasible life-span took over the market. Around the same time, other manufacturers developed a different nuclear cell for pacemakers utilizing the Pm^{147} (direct betavoltaic conversion). Extra precautions were taken in the physical development of the pacemaker resulting in a bulkier and heavier model (DeGraw, 2015). In recent years interest in the technique have been revived due to improved electronics and modern fabrication techniques (Parsonnet *et al*., 2006).

The use as a power source for wireless communication and location tracking sensor has been developed with the creation of ultra-small drone bots. Inspired by the biology of a bee, researchers at the Wyss Institute (Harvard University) are developing RoboBees, manmade systems that could perform myriad roles in agriculture or disaster relief. A RoboBee measures about half the size of a paper clip, weighs less than one-tenth of a gram, and flies using "artificial muscles" compromised of materials that contract when a voltage is applied. Additional modifications allow some models of RoboBee to transition from swimming underwater to flying, as well as "perch" on surfaces using static electricity (Wyss Institute at Harvard University, 2021). Similar devices also being developed by Cornell University, called Reconnaissance Moth. The program's goal is the creation of moths or other insects that have electronic controls implanted inside them, allowing them to be controlled by a remote operator. The animal-machine hybrid will transmit data from mounted sensors, which might include low-grade video and microphones for surveillance or gas sensors for natural-disaster reconnaissance (Lefkowitz, 2019).

Withing the use as a power source for sensors, the current applications are being investigated:

- RFID sensor tag;
- Sensor for location tracking;
- Autonomous Power source of missile encryption module;
- Response to disasters (fire, earthquake, radiation leakage) of nuclear facilities or other sensitive infrastructure without external power supply in case of emergency;
- Aircraft FDR/CVR (black box), ELT (location display device), etc;
- Valves, actuators, marine sensors (oil extraction, pollution, subsea, earthquake detection), telemetry devices, etc;
- Real-time tectonic movement measurement, 4D geological monitoring (Daruich de Souza, 2021; Zhou *et al*., 2021).

CONCLUSION

The push for carbon-free energy that is safe, reliable, and has extra-long duration is high, and nuclear batteries fill all of those criteria.The study and development of betavoltaic batteries is of high interest for science and technology.

Conventional chemical or "galvanic" batteries, like the lithium-ion cells in a laptop or the alkaline batteries, are great at putting out a lot of power for a short amount of time. A lithium-ion battery can only operate for a few hours without a recharge, and after a few years it will have lost a substantial fraction of its charge capacity. Nuclear batteries (among those, betavoltaic cells) by comparison, are produce small amounts of power for a long time. They don't put out enough power for operating a smartphone, for example, but instead can provide a steady little amount of electricity to small devices for thousands of years. In recent years, the demand for autonomous wireless external sensors and with integrated power supply systems has grown. For example, an earthquake sensor that doesn't depend on the electrical power grid lines to work are of extreme interest to society.

Currently, most development projects are in the development/prototype phase linked to research groups at universities and research institutes. In many cases, the process of developing the radioactive source, the semiconductor, and the design are isolated projects carried out by several different researchers within a group or university / institute.

But, even with difficulties, the technology advances. Among the different designs, diamond transducers are preferred because they are the most resistant to radiation. They allow operation over a wide range of temperatures and pressures. New isotopes, such as carbon-14 recovered from nuclear waste (an idea proposed by the University of Bristol in 2016) are also being studied. Monte-Carlo studies are aiding in device development specially when simulating particle quantities and radiation damage.

The major thing one can infer of this field is that an experienced multidisciplinary team with powerfully equipped laboratory/institutes are necessary to designed and develop the next generation of betavoltaic nuclear batteries.

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