High-Energy Dense Power Sources for Low-Power Operation in Extreme Temperature Environments

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Abstract

Radioisotope power sources are being realized in many compelling applications that were deemed unfeasible due to limitations of batteries in temperature, chemical instability, irreversible losses, and electrolyte leakage. Betavoltaics are a rapidly maturing technology that can provide continuous milliwatt to nanowatt power for decades. City Labs' NRC general licensed Nanotritium™ betavoltaic has been demonstrated to be safe, to provide continuous power for over 15 years, and to operate in extreme temperature (-55°C to 150°C) without permanent energy loss or damage, and is available for purchase. Though the technology not a new concept, betavoltaics were discovered in the 1950's, which led to the development of photovoltaics. The development peaked in mid-1970 by Larry Olsen with the Betacel and successfully demonstrated in over pacemaker implants. The growth of the technology has been minimal and sporadic due to lithium battery technology, until recent advancements have materialized in ultra-low power electronics and low-power applications for extreme environments. The Defense Science Board have identified Betavoltaics as a disruptive technology that DOD and NASA should pursue, and now, the technology is being developed for a few applications. Recent work in material development, experiments, modeling, designs, tritium thermoelectrics, and proposed advances will be presented and discussed along with an overview of current projects and improved capabilities. With the development of a lithium-ion battery by Dr. Vilas Pol at Purdue University that can charge and discharge at -100°C, results from a hybrid design with a betavoltaic will be presented. Experimental and modeling will be extended towards commercial and non-commercial applications with near-term advancements and developments to continue the momentum on betavoltaic research.

Keywords

Extreme temperature operation; decadal operating life; solid-state; hybrid battery; NRC General License; tritium; sensor power.

Introduction

Radiation interaction with materials can have beneficial uses, such as in betavoltaic cells, a type of radioisotope power source where the kinetic energy is converted into electricity. Though not a new concept, research and development has been minimal for many years due to limited low-power applications, rapid semi-conductor degradation, limited availability and high cost of suitable radioisotopes, and public perception. Novel and compelling need-based applications are emerging in the military, intelligence, commercial, and medical markets that can utilize the diminutive energy produced from such cells. Present-day sensors, medical implants and electronic devices make betavoltaics an attractive alternative to electrochemical batteries enabling applications to perform for much longer periods in extreme temperatures. Betavoltaics can operate in excess of 10 years over temperatures ranging from -55°C to 150°C.[1] Because the technology is far from mature, many challenges and issues remain to be solved to increase energy density, reduce cost, and make it easier to use. Radioisotopes are used safely in many common applications such as watch dials, gun sights, smoke detectors and exit signs as shown in Figure 1.

Figure 1. Examples of use of radioisotopes

Requirements of the Nuclear Regulatory Commission (NRC) for product licensure often delay development until a sealed source device registry (SSDR) is granted, but recently companies have been working with the NRC and state regulatory bodies to obtain the necessary licensure for distribution of betavoltaic products.

Background

The first betavoltaic battery was developed in 1953 at RCA by Rappaport using strontium-90 (^{90}Sr) but degraded rapidly due to the high beta energy.[2] Others continued research using promethium-147 (^{147}Pm) but were only able to achieve <1% efficiency.[3,4] The most promising development was led by Larry Olsen at the Donald W. Douglas Laboratories in *ca.* 1974 on the Betacel pacemaker battery which exhibited a 4% efficiency using ^{147}Pm and silicon p-n junctions.[5,6] Over 285 patients received Betacel pacemakers, 60 patients inside the United

States. German and U.S. medical institutions were seriously considering the Betacel for wider use but lithium batteries quickly entered the picture and were selected for pacemakers instead.[7] Most advancements in the last 16 years concentrated on designs using tritium with City Labs leading the development by successfully producing NanoTritium™ betavoltaics in 2008 (Figure 2) that are still operating. The betavoltaic design was granted a NRC General License for manufacture and sales within the U.S.

Figure 2. City Labs' NanoTritium™ betavoltaics.

Theory of Operation: Electricity is produced similar to a photovoltaic using the kinetic energy of the beta particles. The basic concept of operation is shown in Figure 3. Beta particles enter the p-n junction and collide with atoms creating electron-hole pairs (EHPs) as they slow down. A 5.7 keV particle, average energy of a tritium beta particle, creates over 1000 EHPs. A portion of the kinetic energy is lost to the lattice. According to the Klein formula, the average kinetic energy required to create an electron-hole pair of energy equal to the semiconductor band gap (E_g) is 2.8 E_g + 0.5 eV while during the conversion process, 1.8 E_g eV and 0.5 eV are lost by emission of acoustic and optical phonons, respectively.[1,8] The voltage developed in a betavoltaic is directly related to the quality of the semiconductor and its band-gap energy.[9]

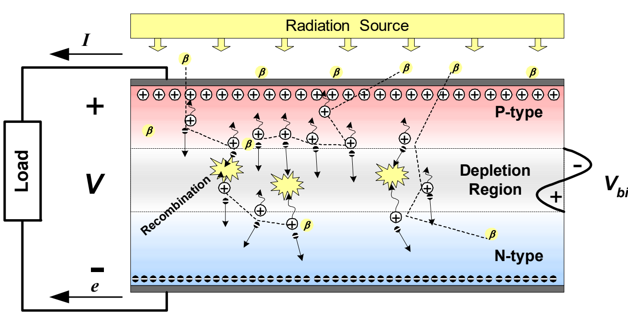

Figure 3. Overview of a betavoltaic cell.

Radioisotope Source: Beta emitting radioisotopes are selected over alpha and gamma emitting ones due to their relatively low mass and short lifetimes. Alpha particles, monoenergetic in the MeV range, have a mass 8,000 times greater than a beta particle immediately damages even the hardest semiconductor materials. Gamma rays are high energy photons (MeV) that travel long distances and difficult to efficiently shield.[10] The most important attributes of beta emitters are the maximum beta energy, decay half-life ($T_{1/2}$), and specific activity (Curies/gram). The high beta energy from ^{90}Sr and krypton-85 (^{85}Kr) exceeds the minimum dislocation energy of 300 - 400 keV for most semiconductor materials which damages the semiconductor and creates Bremsstrahlung.[11] The 25-day half-life of phosphorus-33 (^{33}P) loses 99% of its beginning of life energy (BOL) in 9 months. Therefore, three potential beta emitter candidates are listed in Table 1 with tritium (^{3}H) being the most attractive due to its availability and low cost; readily available from Ontario Power in Canada and is least expensive at \$3.00/Curie (Ci). Russia is the only provider of ^{147}Pm and ^{63}Ni, which are costly at >\$5000/Ci in present day values.[12] Reprocessing in the U.S. would greatly increase the availability.

Table 1. Potential Beta Emitting Radioisotopes.

Isotope	$T_{1/2}$ (yr)	E_{AVG} (keV)	E_{max} (keV)	Ci/g	W/g	$/Ci
^{3}H	12.33	5.7	18.5	9,664	0.33	$3.5
^{63}Ni	100.1	17.1	67	59	0.006	$5k
^{147}Pm	2.6	65	220	600	0.22	* >$5k

** start-up cost is $10M and limited to 200 Ci/year (~$2M/year)*

Designs invoking tritium stored as a tritide demonstrated many practical difficulties in the loading and retention of tritium. Models using MC-SET (Monte Carlo Simulation of Electron Trajectories in solids) were developed to predict surface beta flux for storage materials. Models have been recently developed Monte Carlo N-Particle (MCNP) to continue modeling on other substrate materials such as lithium.

Temperature Performance Results

Betavoltaics were evaluated under temperature from -30°C to 70°C. Current-voltage (I-V) curves were acquired by stepping the applied voltage from 1.00 V to -0.10 V in steps of 1 mV while measuring the current. Figure 4 shows I-V curves from cold to hot, where the open-circuit voltage, V_{oc}, decreased from 0.95V to 0.65V while the short-circuit current (I_{sc}) increased from 82 to 86 nA.

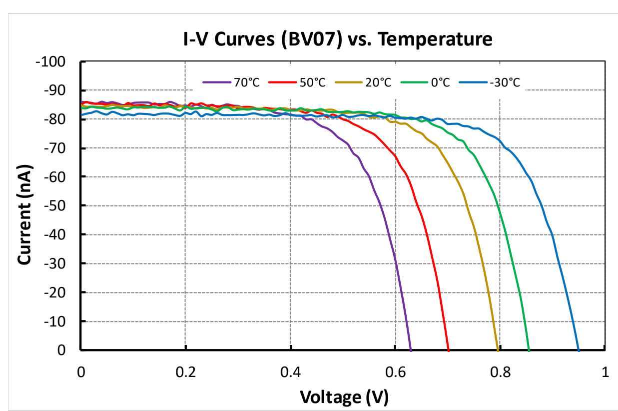

Figure 4. I-V curves of BV07 at -30°C to 70°C

Advances in Higher Power Density

Thinner P-N Semiconductor Substrates: With NASA and Department of Air Force (DAF) driving higher power mission requirements, City Labs has developed process to reduce the thickness of the p-n semiconductor substrate thickness from 625 µm in early P100a devices to <10 µm in new devices (Figure 5). Since tritium betas only travel up to 2 µm, p-in substrates as thin as 5 µm would be sufficient and allow for more betavoltaic layers in a package. Presently, it is possible to stack 50 cells in the NRC general licensed SSD package. The Power Density Roadmap in Figure 6 shows that it is possible to achieve 1 mW/cm^3 or higher.

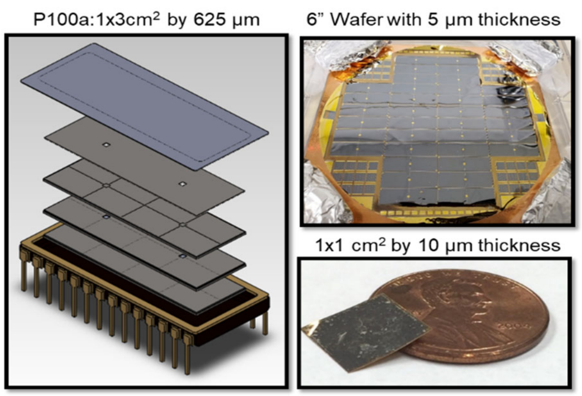

Figure 5. Progression of Thin P-N Substrates

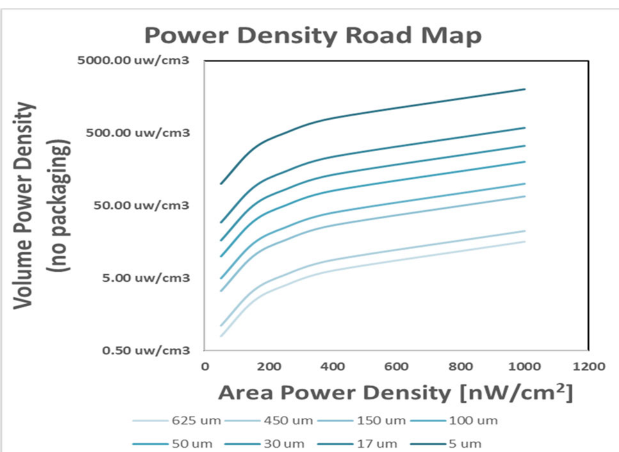

Figure 6. Projected power for Model P200 betavoltaic

Advances in Tritium Source Manufacturing: City Labs has moved into a new facility to establish a state-of-the-art tritium filling capability. The Tritium Filling Rig shown in Figure 7 can handle up to 2.0 MPa (20 bar) of tritium pressures, and temperatures up to 600°C in order to load current hydride materials and new alloys. The tritium capacity of 10g (10 kCi) that can be expanded. The loading process can be automated for production or performed manually to investigate new materials. A new tritium filling vessel was designed to handle the filling rig temperatures and pressures while maintaining an inert atmosphere before extracting loaded films in a glovebox. The combination of the tritium filling rig and vessels provides the only domestic commercial capability to load materials with tritium under high pressure, high temperature, and high activity.

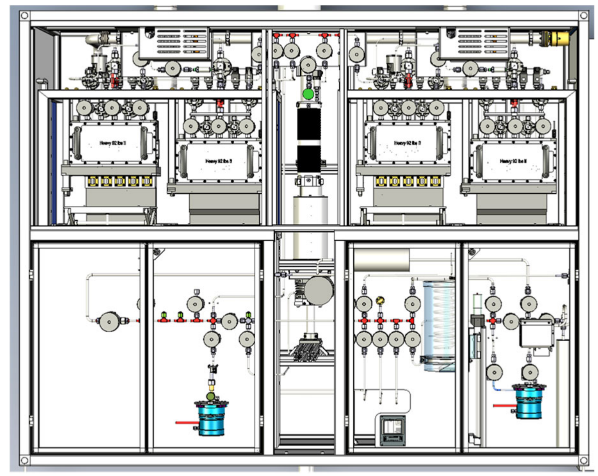

Figure 7. Tritium Filling Rig

Higher Beta Energy Sources: Improvements are being made to increase the tritium density and increase the energy yield from the surface facing the p-n substrate. Preliminary results at Purdue University indicate that lithium films can be loaded safely with hydrogen which would yield higher surface energy as shown in Figure 8. Further development is needed to understand the hydrogen retention behavior. Newer materials such as light-weight high entropy alloys (HEAs) are being investigated.

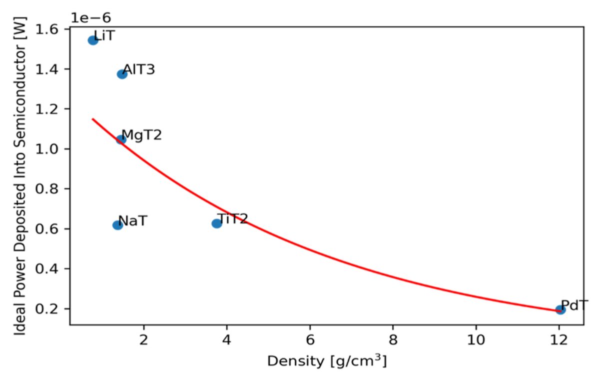

Figure 8. Progression in Tritium Source Substrate

Tritium Thermoelectric Generator (TTG): A novel tritium powered radioisotope thermoelectric generator (RTG) is advancing through two recent research efforts. Prototypes shown in have been developed with deuterium using simulations to estimate the heat power tritium would deliver and project electrical output power.

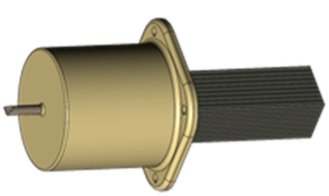

Figure 9. TTG prototype, Patent Pending

There is a point where the tritium-based RTG (TTG) becomes a better potential solution than a betavoltaic. The breakpoint shown Figure 10 occurs at 20 mW (2g or ^3H).

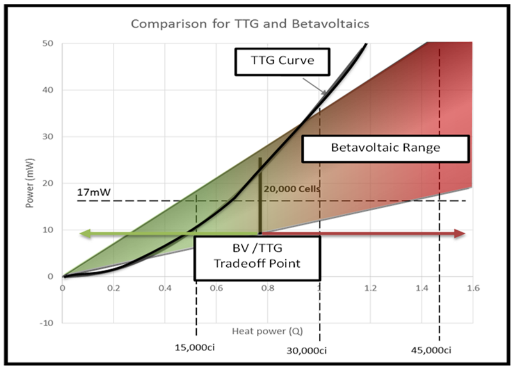

Figure 10. TTG comparison with Betavoltaics

The effects of self-shielding in a TTG are less significant than in a betavoltaic and TTGs more cost-effective in tritium quantities greater than 1-2g. With tritium capable of generating ~0.34 W/g of thermal power, TTGs may present cost and regulatory benefits over traditional RTGs.

Extreme Low Temperature Hybrid Design: Purdue is developing a hybrid battery consisting of a betavoltaic and lithium-ion battery for operation in Arctic and space that can operate below -100°C. To test at temperatures down to -175°C, the Extreme Low Temperature System (ELTS) using liquid nitrogen was developed. The ELTS was validated by cycling a novel lithium-ion cell which received a Guinness World Record for Coldest Operating Battery.[13] The test system is currently being modified for a hybrid betavoltaic and lithium-ion battery demonstration.

Conclusions

Tritium powered devices are a safe enabling technology for DOD, NASA, other government agencies, and commercial applications. Though direct replacement of tritium betavoltaics and RTGs are not straight-forward, work is being done to develop hybrid designs to allow for burst power in extreme temperature environments. Purdue University continues its research in enhancing hydrogen storage quantities in new materials assessing and hydrogen loading system. Purdue is also developing hybrid power sources with their novel lithium-ion battery technology to demonstrate performance at extreme temperatures and beyond. City Labs is leading the development and realization of radioisotope power sources in their betavoltaic and RTG designs.

- Available Supply of Tritium: Department of Energy, National Nuclear Security Administration and Canada Ontario Power Generation
- NRC Licensure: SSDR General, Specific License, Custom Devices
- Transport Regulations: Exempt, Type A, and Type B Packages and Shipment
- Generally Licensed Production & Manufacturing Facility: Achieved Industry's 1st and Only
- City Labs' betavoltaic products not subject to ITAR: 2013 Commodities jurisdiction letter revealed that Models P100a-c and Model p200a-b are not ITAR restricted
- Facility Security Clearance: Achieved Opens Contract Opportunities & Bids on Classified Contracts; Non-active presently

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