Contract No:

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Advancement of Tritium Powered Betavoltaic Battery Systems

The goal of this work is to increase the power output of tritium powered betavoltaic batteries and investigate the change in power output and film resistance in real-time during tritium loading of adsorbent films. To this end, several tritium compatible test vessels with the capability of measuring both the resistivity of a tritium trapping film and the power output of a betavoltaic device *in-situ* have been designed and fabricated using four electrically insulated feedthroughs in tritium compatible load cells.

Energy conversion devices were received from Widetronix, a betavoltaic manufacturing firm based in Ithaca, NY. Thin films were deposited on the devices and capped with palladium to facilitate hydrogen loading. Gold contacts were then deposited on top of the films to allow resistivity measurements of the film during hydrogen loading. Finally, the chips were wire bonded and installed in the test cells. The cells were then baked-out under vacuum and leak checked at temperature to reduce the chances of tritium leaks during loading. Following the bake-out, IV curves were measured to verify no internal wires were compromised, and the cells were delivered to Tritium for loading. Tritium loading is anticipated in October, 2017.

Awards and Recognition

No awards and/or recognitions were received due to participation in this FY15 LDRD project.

Intellectual Property Review

This report has been reviewed by SRNL Legal Counsel for intellectual property considerations and is approved to be publically published in its current form.

SRNL Legal Signature

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Signature **Date**

LDRD-2014-00079
LDRD Report SRNL-STI-2016-00605

Advancement of Tritium Powered Betavoltaic Battery Systems

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Collaborators: Dr. Chris Thomas at Widetronix.

Thrust Area: ST3

Project Type: Strategic

Project Start Date: Febuary 16, 2015
Project End Date: September 30, 2016

Due to their decades-long service life and reliable power output under extreme conditions, betavoltaic batteries offer distinct advantages over traditional chemical batteries, especially in applications where frequent battery replacement is hazardous, or cost prohibitive.

Although many beta emitting isotopes exist, tritium is considered ideal in betavoltaic applications for several reasons: 1) it is a "pure" beta emitter, 2) the beta is not energetic enough to damage the semiconductor, 3) it has a moderately long half-life, and 4) it is readily available. Unfortunately, the widespread application of tritium powered betavoltaics is limited, in part, by their low power output. This research targets improving the power output of betavoltaics by increasing the flux of beta particles to the energy conversion device (the p-n junction) through the use of low Z structured tritium trapping materials.

FY2016 Objectives

- *Complete fabrication and documentation of tritium charging* equipment – This includes assembly, pressure, and leak testing of the remaining tritium exposure vessels, cabling for glovebox feedthroughs, and an instrumentation rack to be used in the Tritium Facilities.
- *Obtain and prepare energy conversion devices* – Obtain energy conversion devices from Widetronix, deposit tritium trapping films, wire bond, and install the devices in the tritium exposure vessels. Bake out under vacuum and leak check the vessels at temperature.
- *Characterize energy conversion devices* – Perform scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS) on films deposited. Measure current voltage (IV) responses for the films with tritium trapping materials deposited.
- *Tritium exposure of films while monitoring resistivity and power output*- Tritium load films while measuring real-time changes in their resistivity to confirm tritium loading. Monitor the power output of the devices after loading is complete.

Introduction

Like all nuclear batteries, betavoltaics convert energy from the decay of a radioactive material into electricity. Unlike other nuclear batteries, however, betavoltaics rely on the kinetic energy of a beta decay rather than the thermal energy (heat) generated by the decay as in thermionics and thermoelectrics. Betavoltaics generate electricity when a semiconducting *p-n* junction is exposed to a beta-particle which in turn excites an electron-hole pair(s) thereby generating an electric current. Conversion of the energy from the beta decay of a radioactive source was initially proposed in the early

1950s. Testing by Rappaport using $^{90}Sr \rightarrow ^{90}Y \rightarrow ^{90}Zr$ not only proved the concept of betavoltaics, but also provided basic theory behind their operation. Unfortunately, the high energy beta emitted by ^{90}Y decay (2.28 MeV) damaged the semiconductor, resulting in a decrease in power output of ~90% over a period of one week. More recent efforts have focused on the use of lower-energy beta emitting materials to minimize long-term damage to the energy collection device. Furthermore, recent research has shown large bandgap semiconducting materials to increase the conversion efficiency of betavoltaic devices [1].

More recent research with tritium has addressed the needs for a low energy beta emitter and for large bandgap semiconductors in the form of SiC *p-n* junctions. However, the flux of beta particles inside the device is still a pressing issue. In the case of tritium betavoltaics this issue has partially been overcome through the use of tritiated thin films due to their ability to store significantly more tritium per unit volume when compared to gaseous tritium as shown in Figure 1.

Isotope	Decay Mode	$T_{1/2}$ (years)	E_{max} (keV)	Activity (Ci/cc)
^{3}H	beta	12.3	18.6	4.53E+06
^{63}Ni	beta	101	66.9	6.30
^{90}Sr	beta	28.8	546	52.2
^{147}Pm	beta	2.6	224	128.8

Figure 1. Potential radionuclides for betavoltaic batteries.

Approach

As mentioned above, increasing the beta flux to the *p-n* junction will increase the power and applicability of betavoltaic devices. The goal of this research is to increase this flux through two avenues: 1) the use of structured tritium trapping materials to increase the amount of tritium stored in the film, and 2) the use of low *Z* tritium trapping materials to decrease self-absorption of the beta particles before they reach the *p-n* junction. Deposition and hydrogenation of structured magnesium films were investigated earlier in the course of this project. An image of a structured magnesium film is shown in Figure 2. Tritium loading of magnesium films was not performed.

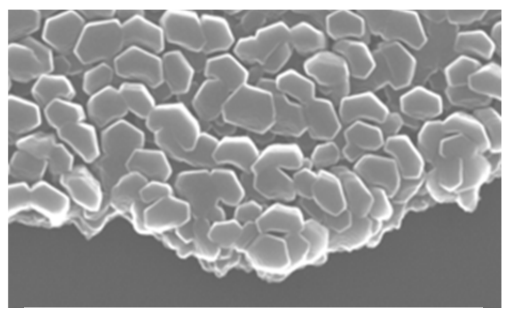

Figure 2. Structured magnesium film

Traditionally, pressure, volume, and temperature (PVT) measurements have been used to verify and monitor the hydrogenation/tritiation of a hydride under a given set of conditions. Because only minute amounts of gas are absorbed by the thin films used in betavoltaics, an alternate method was needed. The test cell shown in Figure 3 is equipped to monitor the resistivity of the film in real-time during the loading process. The films transition from metallic conductors to insulators as hydrogen is absorbed. Additionally, the tritium loading cell provides the capability to monitor the power output of an experimental device without removal from the cell.

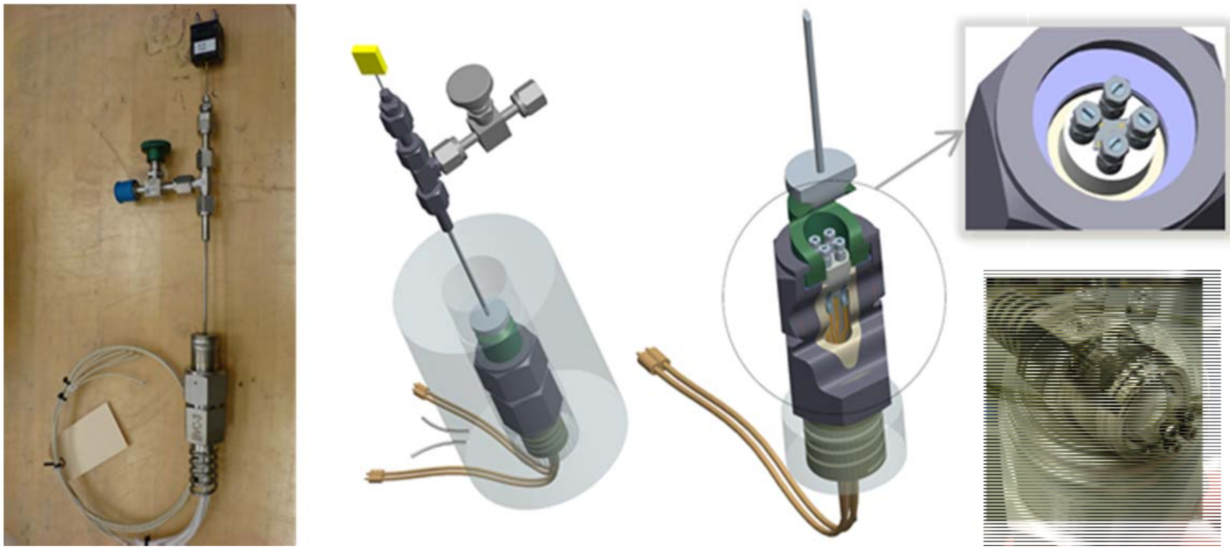

Figure 3. Tritium loading cell designed and fabricated at SRNL.

Results/Discussion

Betavoltaics received exploratory funding in February of FY16 to complete tritium loading. Final pressure and leak testing of 8 test vessels were performed (including rework) as well as completion of the data acquisition system and associated cabling. Energy conversion devices were received from Widetronix, a betavoltaic manufacturing firm based in Ithaca, NY. The devices had been produced to deposit gold contacts for power measurements. Once the contacts were deposited, photoresist was applied, exposed to UV light. The films were then shipped to SRNL where the photoresist was developed to expose a fresh surface to deposit tritium trapping films. Thin films of titanium were then deposited on the device and capped with palladium. Gold contacts were deposited in the corners of the films before the remaining photoresist was removed. These steps are shown graphically in Figure 4. Removal of the remaining photoresist exposed the gold power contacts, which were wire bonded, installed in the test cells, and IV (current-voltage) curves were measured. The cells were then baked-out under vacuum and leak checked at temperature to reduce the chances of tritium leaks during loading. Following the bake-out, the IV curves were re-checked to verify no internal wires were compromised, and the cells were delivered to Tritium. Figure 5 shows a representative graph of the Widetronix device measured after mounted in the load cell and bake out. The figure shown to the right is the data for devices currently in Tritium Facilities for future resistance and power measurements. The data was taken using the electronics that will be utilized for the betavoltaic measurements in Tritium. The software written for the electronics was tested with a commercially available solar cell. Figure 6 displays a representative data set for the solar cell and includes the power versus time plots from four simultaneously measured solar cells in real time. A power drop is seen in the SMU2B channel after covering part of the associated solar cell. The identified open circuit voltage (V_{oc}) is the maximum voltage of the solar cell at which the net current is zero.

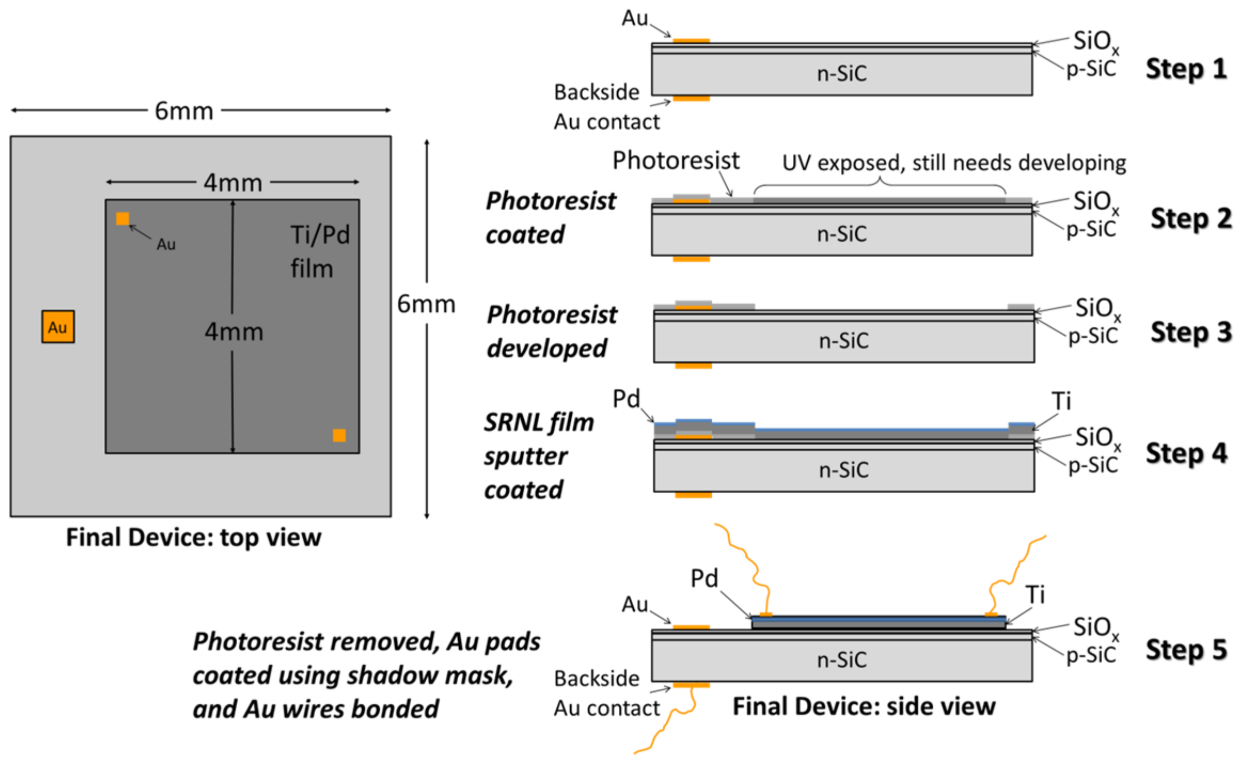

Figure 4. Schematic showing steps taken to deposit SRNL films on top of Widetronix devices.

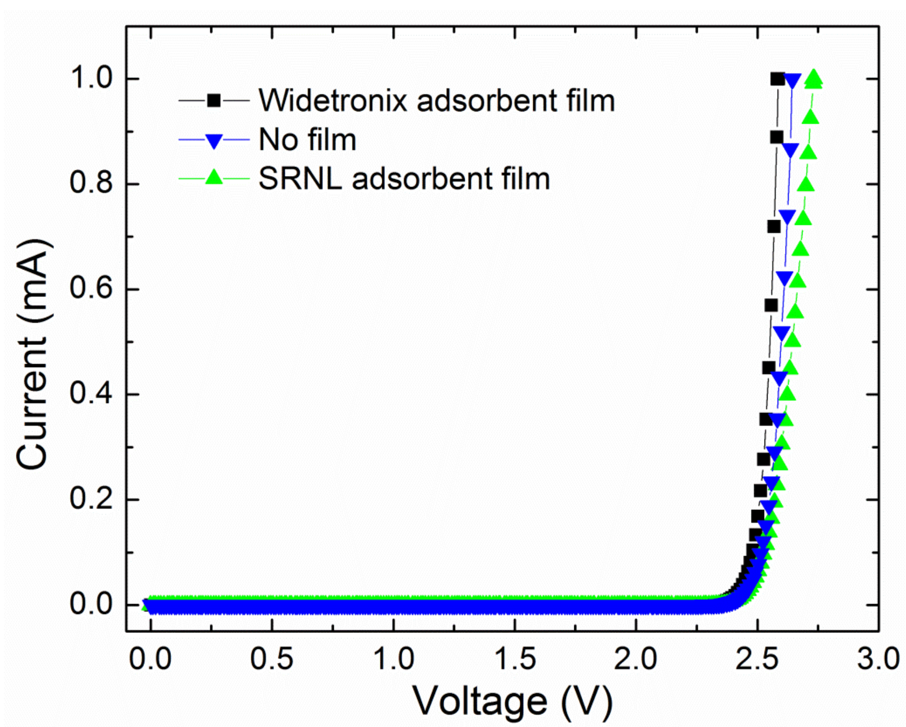

Figure 5. IV curve for Widetronix device measured in SRNL load cell. Insert shows log plot of device.

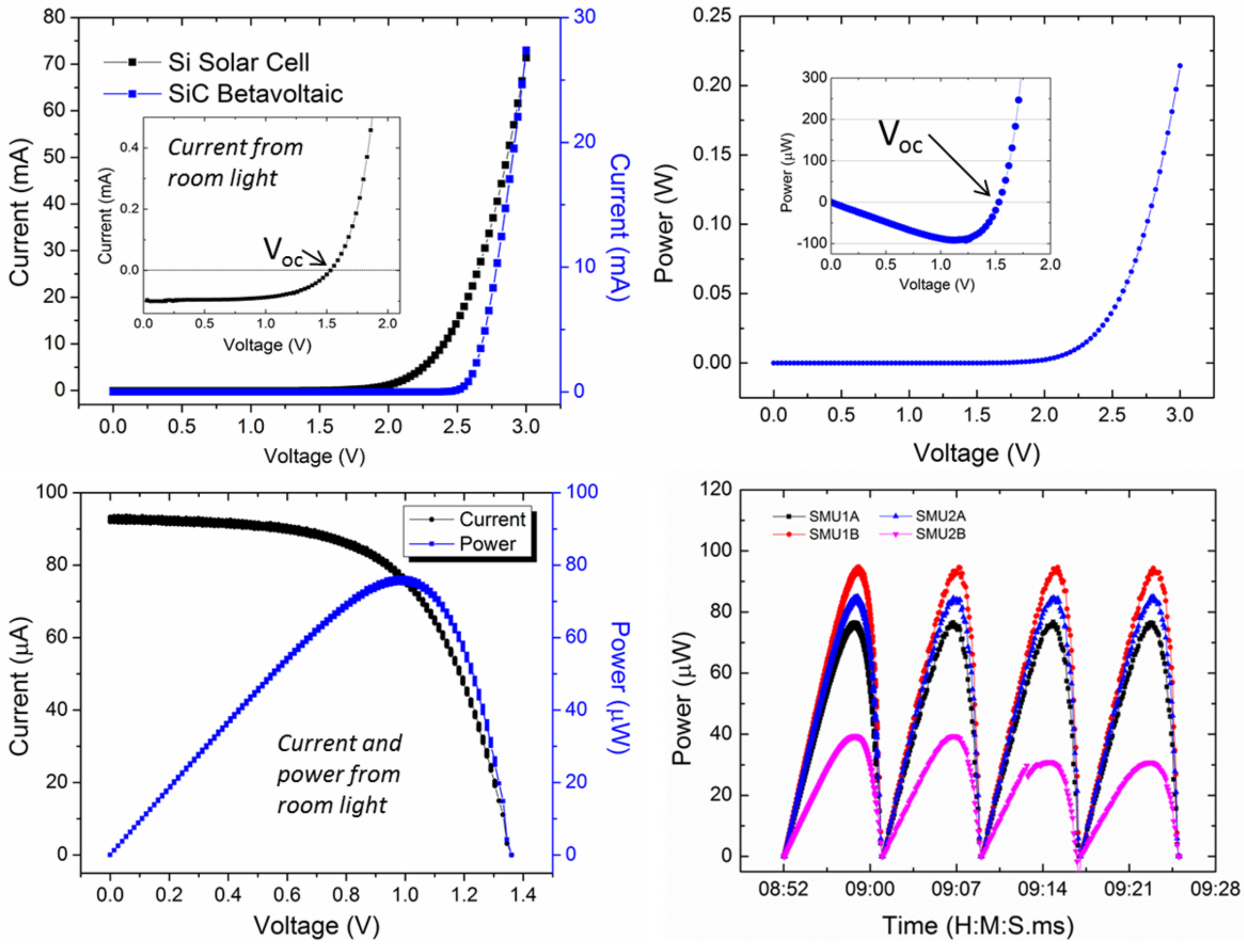

Figure 6. Solar cell data taken with electronics built for real time testing of betavoltaic devices during tritium uptake.

Tritium loading of the betavoltaic devices did not occur this fiscal year. This is primarily due to three reasons: competing facility priorities, delays associated with fabrication of cabling feedthroughs, and rupture disc challenges. Loading of the first four samples is currently scheduled for this October. Once loading of the first sample set has been achieved and capabilities have been demonstrated, additional funding may be pursued.

FY2016 Accomplishments

- Completed Task Technical Plan permitting loading in the SRTE
- Completed pressure and leak testing on nine loading cells (included rework)
- Completed fabrication of cabling needed for tritium loading
- Received energy conversion devices from our corporate partner, Widetronix
- Deposited Pd capped Ti films using OAD in a sputtering chamber at SRNL
- Leak checked and baked out films installed in loading cells
- Verified loading cell functionality by obtained IV curves on films deposited in loading cells
- Delivered loading cells and all needed equipment to SRTE for tritium loading

Future Directions

- Tritium loading of films is anticipated in FY17.
- Verification of tritium load results and cell measurement capability will allow pursuit of additional funding.

FY 2016 Publications/Presentations

No external publications or presentations have been made to date.

References

1. L. C. Olsen, "Review of Betavoltaic Energy Conversion," in Proc. 12th Space Photovolt. Res. Technol. Conf, 1993, p. 256.

Acronyms

HRTL: Hydrogen Research and Technology Lab
OAD: Oblique Angle Deposition
PVT: pressure, volume, and temperature
SRNL: Savannah River National Laboratory
SRTE: Savannah River Tritium Enterprise

Intellectual Property

Invention disclosure has been drafted.

Total Number of Post-Doctoral Researchers

One postdoctoral researcher (Timothy Krentz)
One summer intern (Ian Demass, Clemson)