Contract No:

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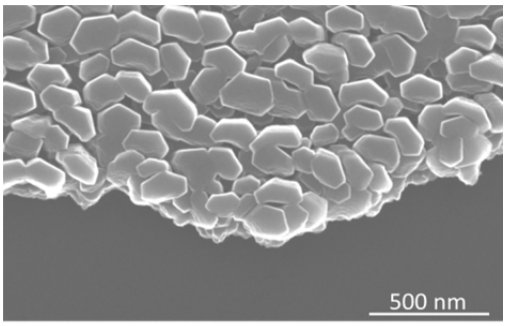
Disclaimer:

Advancement of Tritium Powered Betavoltaic Battery Systems

The goal of this work is to advance tritium powered betavoltaic battery systems. To this end, we have developed and fabricated several tritium compatible test vessels that have the capability of measuring both the resistivity of the tritium trapping film and the power output of a betavoltaic device in situ. These polymer-free vessels use a ceramic feed-through to provide the electrical insulation needed for resistivity and power measurements of the film and device, respectively. These measurements can be made in tritium at temperatures up to 450 °C and 950 psig. An early prototype of the vessel was used to demonstrate hydrogen loading of novel palladium capped nano-structured materials.

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 nanostructured tritium trapping materials. With this in mind we have grown and hydrogenated nanostructured magnesium films.



Microstructured Mg film (Pd capped, ~5nm) grown using Oblique Angle Deposition (OAD).

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

Signature **Date**

LDRD-2014-00079
LDRD Report SRNL-STI-2015-00544

Advancement of Tritium Powered Betavoltaic Battery Systems

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Collaborators: Prof. Timothy DeVol at Clemson University and Dr. Chris Thomas at Widetronics.

Thrust Area: ST3

Project Type: Strategic

Project Start Date: October 1, 2013
Project End Date: September 30, 2015

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 nanostructured tritium trapping materials.

FY2015 Objectives

- *Optimize hydride films* –Optimization of hydrogen pressure, material temperatures, and loading times required to create hydrogenated films while maintaining their integrity. Produce next generation tritium loaded films that would maximize tritium loading while minimizing beta particle self-absorption.
- *Develop a tritium charging vessel* – Completion of the design and fabrication of several tritium certified vessels. These vessels are novel in that they will allow measurements to be made of both the resistivity of the films and power output of the devices in real time during tritium loading at temperatures up to 450 °C and pressures up to 950 PSIG.
- *Tritium exposure of films while monitoring resistivity and power output*- Tritium loading of films using the optimized hydride parameters while measuring real-time changes in resistivity to confirm tritium loading, and monitoring the power output of the device after the loading is complete.
- *Quantification of tritium loading* – Quantification of tritium loading using liquid scintillation and digestion.

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. A simple schematic of this setup is shown in Figure 1.

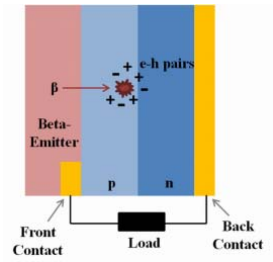

Figure 1. Diagram of a typical betavoltaic cell.

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. [1]

Approach

As mentioned above, the main hurdle for the widespread implementation of tritium betavoltaics is the low beta particle flux delivered to the *p-n* junction in current devices. The goal of this research is to increase this flux through two avenues: 1) the use of nanostructured 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.

Traditionally, pressure, volume, and temperature (PVT) measurements have been used to verify and monitor the hydrogenation/tritiation of a material under a given set of conditions. Because only minute amounts of gas are absorbed by thin films, an alternate method was developed. The test cell shown in Figure 2 is equipped to monitor the resistivity of the film real-time during the loading process. Resistivity is monitored because the films transition from metallic conductors to insulators as loading progresses. Additionally, the electrical feedthroughs in the tritium loading cell provide the capability to monitor the power output of an experimental device without removing it from the cell. To confirm the loading conditions, a test station was also designed and built at the Hydrogen Research and Technology Lab (HRTL) to load samples with protium in order to demonstrate that the loading technique was sound before initiating testing with tritium.

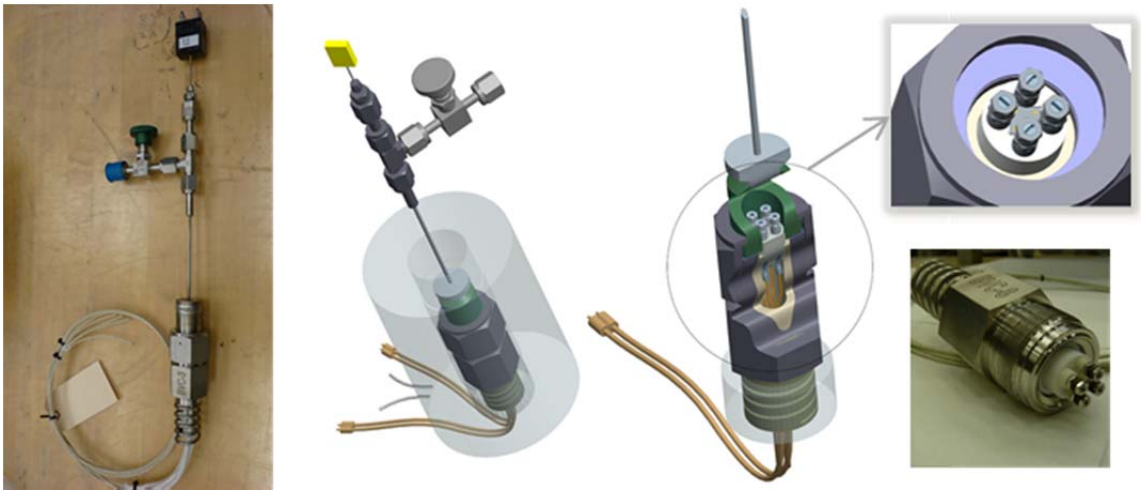

Figure 2. Tritium loading cell designed and fabricated at SRNL.

Results/Discussion

In FY15, palladium-capped magnesium thin films were deposited using traditional rotating substrate sputtering. In addition, nanostructured palladium capped magnesium thin films were deposited using Oblique Angle sputtering Deposition (OAD). The OAD films, shown in Figure 3, were grown in a sputter deposition chamber with a sample-to-source angle of ~85°. The goal of the nanostructured magnesium films was to increase the mass of tritium stored in a given volume by increasing the surface area of the film, thereby allowing more complete hydrogenation.

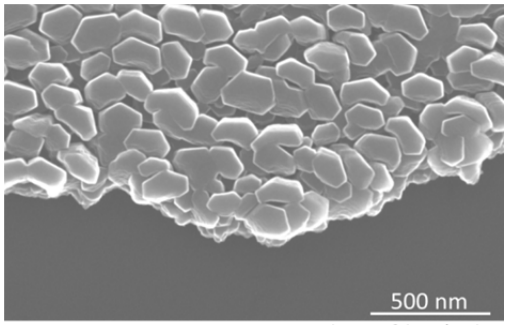

Figure 3. Microstructured Mg film (Pd capped, ~5nm) grown using Oblique Angle Deposition (OAD).

A test station was also built at HRTL to test the protium loading of thin films in order to evaluate the appropriate PVT conditions for tritium loading. A number of experiments were performed in which the resistance of the film was monitored during the hydrogen loading process under different conditions. Figure 4 shows a sample resistivity (ρ) versus time plot for a loading sequence. Arrows indicate changes in reaction rate (the slope of the ρ *versus T* plot) that correspond to changes in the H_2 pressure (20-50-150 psi) in the chamber. As expected, the H_2 loading rate increased with increased pressure.

In FY15, the tritium loading cell design that met tritium requirements related to materials, pressure protection and welding, was finalized. A supporting ASME code calculation was performed (Design temperature: 450 °C and Design Pressure: 950 psig) and Record Drawings for Tritium Facility installation within the glovebox were developed. Certification and approval of the electrical feedthroughs in the cell were particularly important. In order to achieve the desired pressures and temperatures a Ceramtec™ fitting was chosen. This choice was key to the project as it supported a small cell size that facilitates favorable process heat transfer conditions. The fitting contains a complex mix of metal, ceramic, and braze alloy making code extrapolation of unheated pressure test results particularly challenging. Therefore, the component had to be pressure and leak tested at the operating temperature of 450 °C. This was the first heated leak/pressure test completed in the SRNL High Pressure lab. Figure 2 shows an assembled certified tritium loading cell. Currently three cells are ready to be deployed in the tritium facility (assembled and pressure/leak certified) with a further 9 cells assembled awaiting pressure and leak certification.

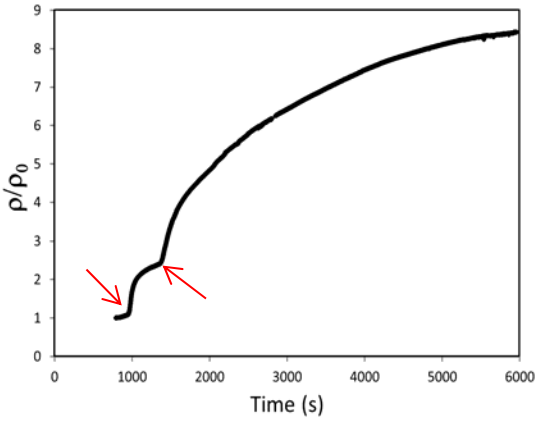

Figure 4. Normalized resistivity as a function of time during hydrogen loading of a Pd capped Mg film.

FY2015 Accomplishments

- Grew nanostructured Mg films capped with Pd using OAD in a sputtering chamber at SRNL.
- Built a test station for protium loading thin films in an environment similar to the tritium facility
- Protium loaded Pd capped Mg films grown at SRNL
- Completed design and certification of a tritium approved loading cell with the capability of monitoring electrical properties real-time.
- Completed fabrication of 12 loading cells
- Completed pressure and leak testing on three loading cells

Future Directions

- Tritium loading of films is anticipated in FY16, contingent upon receipt of additional funding.

FY 2015 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

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

Intellectual Property

Invention disclosure has been drafted.

Total Number of Post-Doctoral Researchers

One postdoctoral researcher (Dale Hitchcock), and one summer intern (Ian Demass, Clemson) were involved with this project.