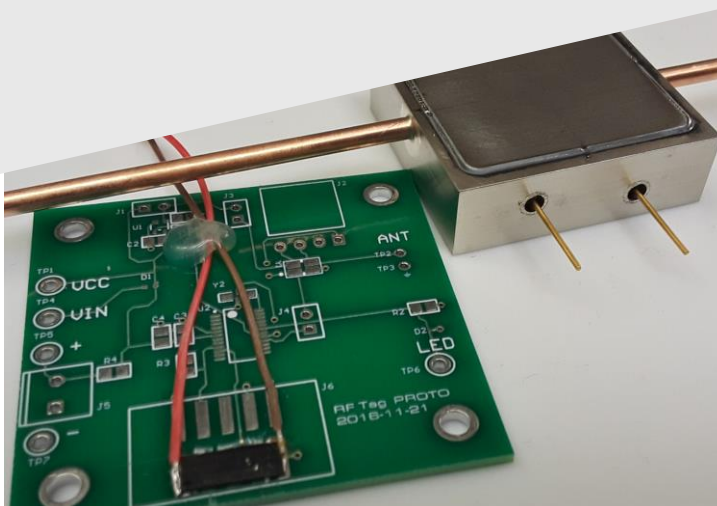


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Tritium Betavoltaic Powered Sensor Platforms: Power Augmentation with Scintillating Particles

September 2021

Lance Hubbard

Erin Fuller

Richard Lundy

Yadexy Rodriguez

Sarah Mendoza

Zachary Kennedy

Jarrold Allred

Radha Motkuri

Gary Sevigny

Rebecca Erikson

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Pacific Northwest National Laboratory
Richland, Washington 99354

Abstract

Betavoltaics (BV) are long-life power sources that typically convert beta particle radiation into electricity. Largely, the radioactive decays within the source go unharvested by the device. This work seeks to augment the power generation of BV devices by integration of scintillating particles within the radiative getter to convert beta emission which would otherwise not leave the getter into usable light for power generation. Silica-covered barium fluoride scintillating particles were integrated into a tritiated water getter. Power generation was increased from 100s of nW to μ W levels with the addition of 0.2 wt. % particles into the getter. Nanowatt-scale sensor platforms were demonstrated with the μ W BV devices and the maximum possible lifetime of such platforms was estimated. As this technique enables higher power density BV devices from conventional Si semiconductors (compared to wider bandgap BVs), the further implementation may lower the barrier-to-deployment of these long-life power/sensor platforms.

Acknowledgments

The authors would like to acknowledge Michelle Fenn for her assistance with the additive manufacturing of the chemical sensor. This research was conducted under the Laboratory Directed Research and Development Program at Pacific Northwest National Laboratory, a multiprogram national laboratory operated by Battelle for the U.S. Department of Energy.

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1.0 Introduction

Betavoltaics (BV) are long-life power sources that typically convert beta-particle radiation into electricity (L. C. Olsen et al., 2012) by way of thin-film two-dimensional stacks of radioactive material and semiconductors (Alam & Pierson, 2016). Currently, BV designs are separate from beta photovoltaics (BPV) which first turn the energy from the beta particles to photons through means of a scintillator placed between the radiation source and the semiconductor (Berman et al., 2018; Russo et al., 2017; Yuruk & Tutunculer, 2017). Both designs produce electricity only from the radiation emitted from the source, which accounts for a fraction of the total radioactive decays (Hubbard, Cowles, et al., 2020; Spencer & Alam, 2019; Zaitsev et al., 2014). Some augmentation of device power efficiency has been done by the design of three-dimensional BVs (Murphy et al., 2019) which incorporate complex pores and structures into BV designs. Largely, the radioactive decays within the source go unharvested by the BV/BPV device.

This work seeks to augment the power generation of a BV device by integrating scintillating particles within the radiative getter to convert beta emission which would otherwise not leave the getter into usable light for power generation. In this way, some of the energy of beta decays that would have been lost can be harvested. The integration of scintillators into the radioactive source effectively blends BV and BPV designs to present a more efficient and elegant design.

This work describes the integration of scintillating particles into the tritium getter matrix of p-n junction silicon-based betavoltaics to enhance device performance. Example nanowatt-scale electronic sensor platforms are powered with betavoltaics as an initial demonstration and to gain some insight into the expected lifetime of such platforms. Specifically, we hypothesized that:

- The addition of silica-covered barium fluoride particles (5-100 micron diameters, 100 nm thick silica shell) into the tritium getter matrix with concentrations ranging from 0.18 to 1.77 mg/ml increases the optical light available for power generation.
- That the use of a betavoltaic with integrated scintillators into the getter matrix can power sensors platforms needing 375nA and below.

As this is the first time, to our knowledge, such integration of scintillating particles has been attempted, an important parameter of interest is the effect of particle concentration on the energy output. As an example of performance, nanowatt-scale electronics are powered with the BVs, and the augmented BVs enable many more possible years of platform life compared to the control case. The integration of scintillating particles into a radioactive source enables the augmentation of BVs based on more conventional (and available) semiconductors. It is hoped that this augmentation path will reduce the barrier-to-deployment of BV technology.

2.0 Experimental

The getters were composed of a mixture of superabsorbent polymer (SAP), polymer stabilizers, metal-organic frameworks (MOF), and scintillating particles. This transparent getter was used to demonstrate the efficacy of incorporating scintillating particles into the radiative matrix. The stabilizers and MOF were added to boost the longevity of the SAP under beta irradiation, and the scintillator concentration was the variable of the experiment. The synthesis procedures for the individual getter components follow.

2.1 Materials

Poly(acrylic acid) partial potassium salt (PACA-K, CAS Number: 25608-12-2), barium fluoride powder (BaF_2 , 99.999+%, CAS Number: 7787-32-8), tetraethyl orthosilicate (TEOS, CAS Number: 78-10-4), sodium polyacrylate (PA, CAS Number: 9003-04-7), fumaric acid (CAS Number: 110-17-8), formic acid (CAS Number: 64-18-6), and zirconyl chloride octahydrate ($\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$, CAS Number: 13520-92-8) were purchased from Millipore-Sigma and used without further purification. All chemicals were of reagent grade (95+%, except the barium fluoride which was 99.999+%). All solvents were acquired from PNNL dry storage and used without further purification.

2.2 Scintillator Particle Preparation

The silica-covered barium fluoride was produced by grinding barium fluoride powder in an agate pestle and mortar for 15 min until a very fine powder is achieved, which sticks to the side of the smooth wall. The powder was then silica covered using the same procedure as was previously reported (Hubbard, Sumner, et al., 2020) with the scintillator particles being used instead of the quantum dots. The glass-covered powder was collected by centrifugation and then washed by five alternating cycles of dispersion and centrifugation. A very fine silica-covered powder was collected and vacuum dried for 24 hours to remove any remaining reagents.

The particles were characterized by photoluminescence imaging on an OMAX 2500 EPI-fluoroscope. The nominal particle size was found to be $5.3 \pm 0.5 \mu\text{m}$ with some (<10%vol) larger particles with diameters larger than $100 \mu\text{m}$. These particles were stored under Ar at ambient temperatures until integration into the getter.

2.3 Metal-Organic Framework Preparation

Metal-organic framework particles were added to stabilize the getter matrix. Traditionally used water sorbent materials such as metal salts, silicas, zeolites, and carbons have limited porosity and affinity, resulting in low sorption capacities. In this direction, newer synthetic sorbents such as MOFs have been exploited for water adsorption, mainly because of their tunability, porosity, and high surface areas (Figure 1a) (Liu et al., 2018; Ramesh et al., 2009; Zheng et al., 2017, 2020). Further, these materials may be tailored for enhanced sorption capacities and high stability to radiation. To this end, we considered zirconium-based structures because of their high stabilities and hydrophilic character. Zr-hydroxo clusters with strong Zr-O coordination bonds and a high degree of 12-fold connectivity clusters (Figure 1) provide very high stability (i.e., to elevated temperatures) and are chemically resistant. The zirconium MOF-801 with a very short linker (fumaric acid) has shown exceptional water sorption characteristics at low partial pressures. Using this modulation approach, we synthesized a naturally occurring unbranched unsaturated dicarboxylic acid with zirconium oxychloride salt in acidified

dimethylformamide (DMF) solvent at 130 °C. After cooling, the white precipitate was washed with 10 mL DMF followed by 10 mL ethanol and dried in air.

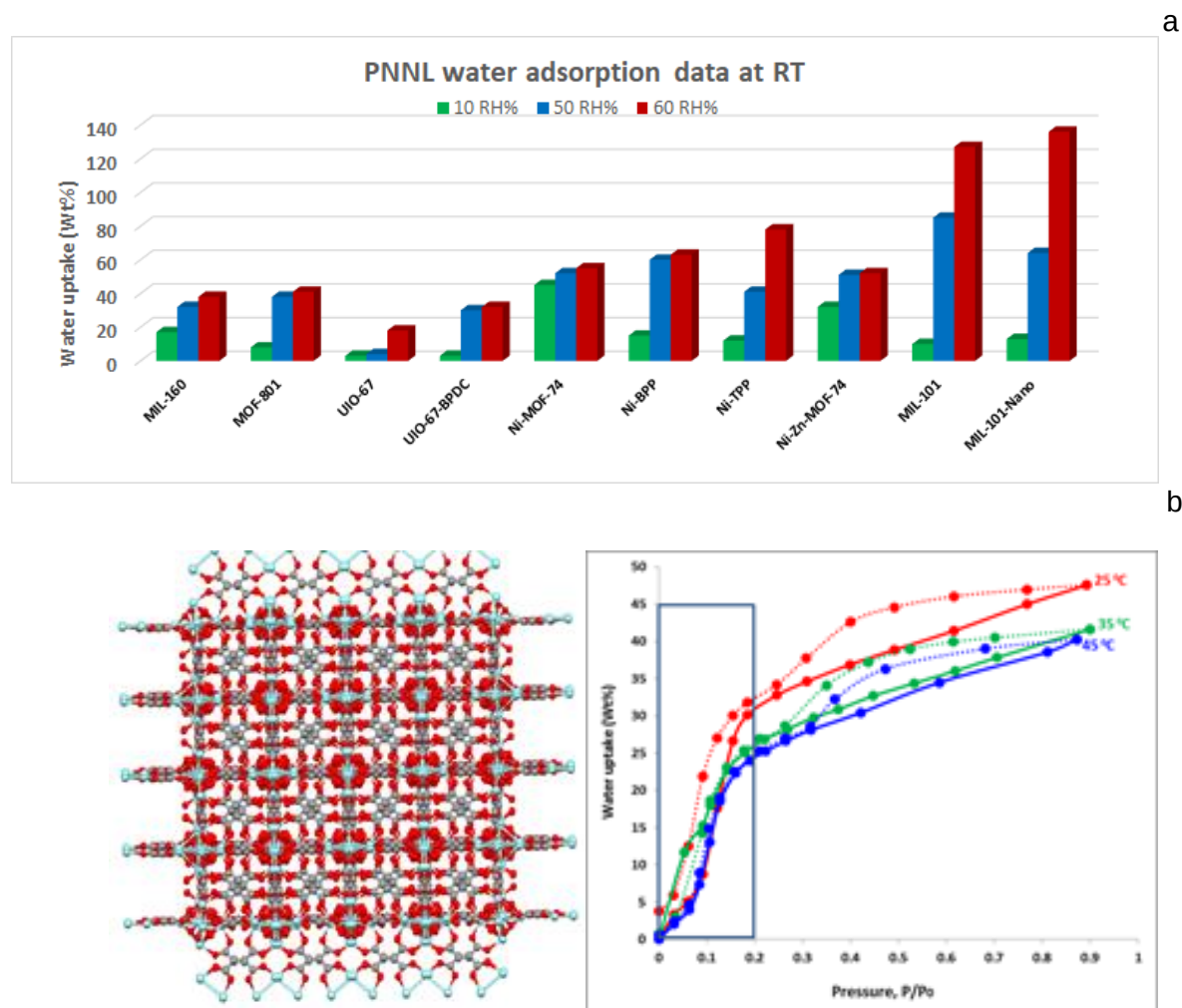


Figure 1. a) Water adsorption characteristics of a few MOFs at three different RH conditions (10, 50, and 60%) and b) MOF-801 crystal structure and its water adsorption performance at three different temperatures

Powder XRD measurements showed the crystallinity of the MOF sample with a pattern matching previous literature reports (Zheng et al., 2017). To test the water sorption capacity, the MOF sample was further activated at 150 °C for 12 hours under a nitrogen atmosphere to remove any trapped solvent molecules from the pores. The surface area analysis and pore-size distribution studies were performed to determine the pore characteristics. The water adsorption measurements were performed using a batch gravimetric sorption instrument. It is exciting to note that the MOF-801 showed the best performance during water sorption measurements, ~30 wt% at 6 millibars and 35 wt% at 10 millibar pressure at room temperature (~32 wt% at P/P_0 of 0.2 at 25 °C, reaching 48 % at P/P_0 of 0.95). Because of its water adsorption characteristics, the MOF-801 was further tested for water sorption experiments at various temperatures. The MOF-801 sample was activated at 200 °C for 12 hours under a vacuum to remove any trapped solvent molecules from the pores, specifically methanol and any trace DMF molecules. The

water sorption experiments were performed at 35 °C and 45 °C, and the sorption behavior was plotted against the partial pressures, as shown in Figure 1. From the sorption behavior, it is clear that the sorption capacities were maintained even at higher temperatures. The desorption curve goes all the way to zero, which shows a very good lift in the sorption capacity along with good recyclability of the material. It is interesting to note that, just by applying a simple vacuum, the sample returned to its original weight, which is an essential property of the material for better recyclability.

MOF-801 was successfully synthesized and tested on a 100-gram scale (Zheng et al., 2017). The synthetic procedure, briefly, involved 50 mmol of each fumaric acid and $\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$ were dissolved 500 mL screw-capped jar in a mixed solvent of DMF and formic acid (200 mL and 70 mL) and was then heated at 130 °C overnight to give a white precipice, MOF-801. The product showed excellent crystal structure as well as sorption characteristics.

2.4 Betavoltaic Preparation

The getters were a mixture of deionized water, PA, MOF 801, and scintillating particles. Using a doctor blade deposition process, the getter films were applied to the active face of the semiconductors with a uniform thickness. The super absorbent polymer was chosen for the main getter component, as it allows for the maximum amount of radioactive water to be stabilized per unit volume, which is important to harvest the most power out of the device. As the polymer is not anticipated to last the full lifetime of the device, this getter choice is temporary to facilitate testing of the scintillating particles. The getters had the consistency of a gel when applied.

The chips were dried under a low (< 1 torr) vacuum for 24 hours to remove any of the original water and prepare the getter to receive the tritiated water. The dried chips were then prepared to be connected to the BV boxes. The final getter compositions of each betavoltaic are presented below:

Table 1. Tritiated Water Getter Compositions

Weight Percent:		Paca-K	MOF 801	Scintillator Particles	PA	Water
Betavoltaic:	1	0.20%	0.22%	0.00%	0.02%	99.56%
	2	0.20%	0.22%	0.02%	0.02%	99.55%
	3	0.20%	0.22%	0.04%	0.02%	99.52%
	4	0.20%	0.22%	0.09%	0.02%	99.48%
	5	0.20%	0.22%	0.18%	0.02%	99.39%

The containment boxes for the betavoltaics were manufactured by Hermetic Solutions Inc. The boxes contained space for the internal components, brazed copper lines for radioactive gas introduction, and two hermetically isolated electrical lines for power transmittance. The outer dimensions were 36.8 mm X 35.5 mm X 17.1 mm, with internal volume measuring approximately 25.4 mm X 25.4 mm X 15.0 mm. A 0.5 mm thick lid was provided and laser/stipple tungsten arc (TIG) welded after the boxes were populated.

The BVs were internally populated with Aoshike 19 mm X 39 mm unencapsulated polysilicon solar cells cut to fit into the box. Five cells were connected in series with aluminum tabbing and conductive epoxy. The cells were then soldered to the I/O leads for the box by 150 um diameter

platinum wire. Figure 2a shows a side view diagram of the stacking order and the relation of various pieces to one another. Figure 2b shows a top-down view of the chips inside the box before the soldering of the final Pt wire between the chip stack and the power transmission lead.

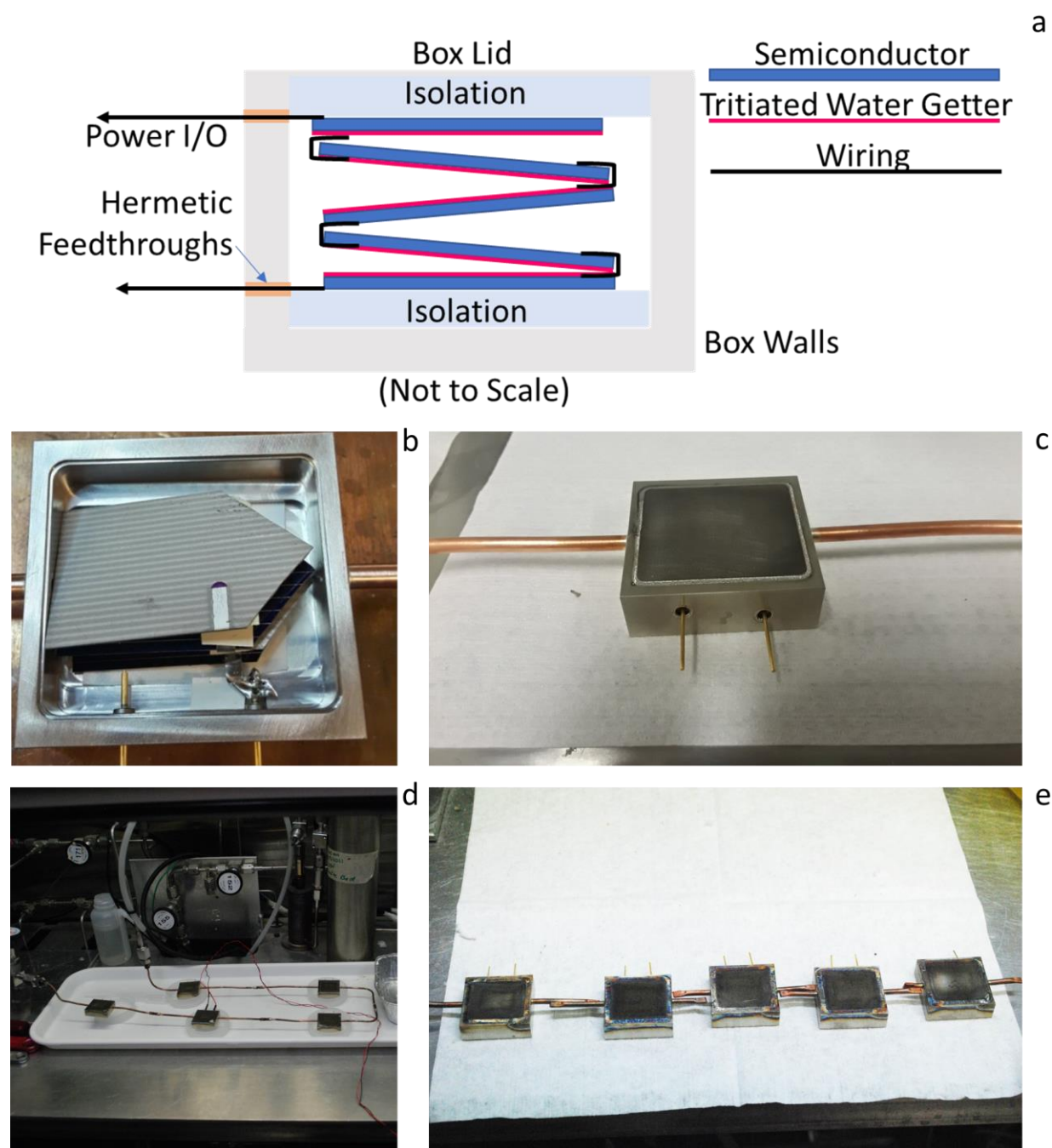


Figure 2. a) Stack order diagram of the internal components of the betavoltaics. b) Photograph of the internal components of the betavoltaics before final terminal connection and welding. c) Betavoltaic after laser welding closed box. d) The BVs welded together into a line for tritiated water introduction. e) The BVs after cold welding and cutting of the gas lines.

After the box population, the lid was initially welded on by laser welding with an Innovative Laser Technologies Glovebox laser processing system at 300W power. An example sealed BV is

seen in Figure 2C. The welds were tested with 5 psi He leak checks, and stippled TIG welding was employed to seal any leaks in the weld. When the 5 BVs were verified to be sealed, they were welded together into a linear arrangement by gas leads (Figure 2d), rechecked for leaks, and then connected to the tritium loading manifold by quarter-inch Swagelok fittings.

2.5 Tritiated Water Introduction

Source gas used to load the betavoltaic cells was procured in 2007 as part of a 900 Ci tritium gas order from American Radiolabeled Chemicals Inc. Gas composition analysis via Finnigan MAT-271 magnetic sector mass spectrometer (see Figure 3) determined that the gas was 23.9 mol% tritium gas (see Table 2), correlating to 119 Ci.

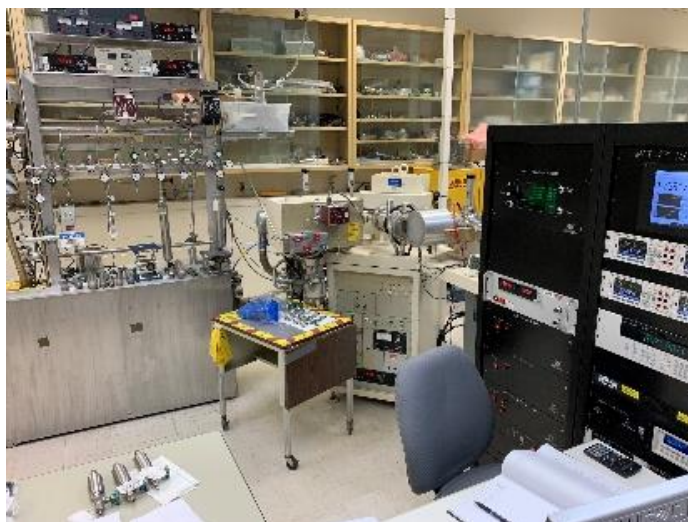


Figure 3. Finnigan Mass Spectrometer

Table 2. Source Gas Composition

Component	Mol %		Estimate Uncertainty
Hydrogen	0.069	±	0.001
He-3	73.8	±	0.2
T2	23.9	±	0.3
N-28	2.19	±	0.04
Argon	0.038	±	0.02
Total	100		

A copper oxide bed was used to convert the tritium gas to tritiated water, which could then be absorbed onto the betavoltaic cells. This copper oxide bed was heated to 600 °C and evacuated for an hour to remove any potentially contaminating off-gases. The bed was isolated from the manifold and the pressure was monitored to ensure all off-gases were removed. The transfer gas manifold and betavoltaic cells were then pumped down to 150 mtorr. From there, the manifold valves were aligned to isolate the vacuum manifold from the BV portion as seen in Figure 4. Gas was then slowly introduced from the source gas bulb to the copper oxide bed and following BV cells. Once pressure equilibrated the back-end valve of the BV portion of the manifold was opened and the source gas could slowly expand into additional portions of the

evacuated transfer manifold. As there was a large portion of He-3 in the source gas it was important to flow this gas over the cells slowly to facilitate more T-2 reaching the cells. Ice was then added to the top and bottom of the betavoltaic cells to promote tritiated gas to condense at those locations. With these expansions, 31 Ci was left in the source vessel, so 88 Ci could have been in contact with the BV cells. The source gas vessel was closed and the manifold was backfilled with ultra-high purity argon to 650 torr. The valves to isolate the BV cells were closed and the rest of the manifold was evacuated.

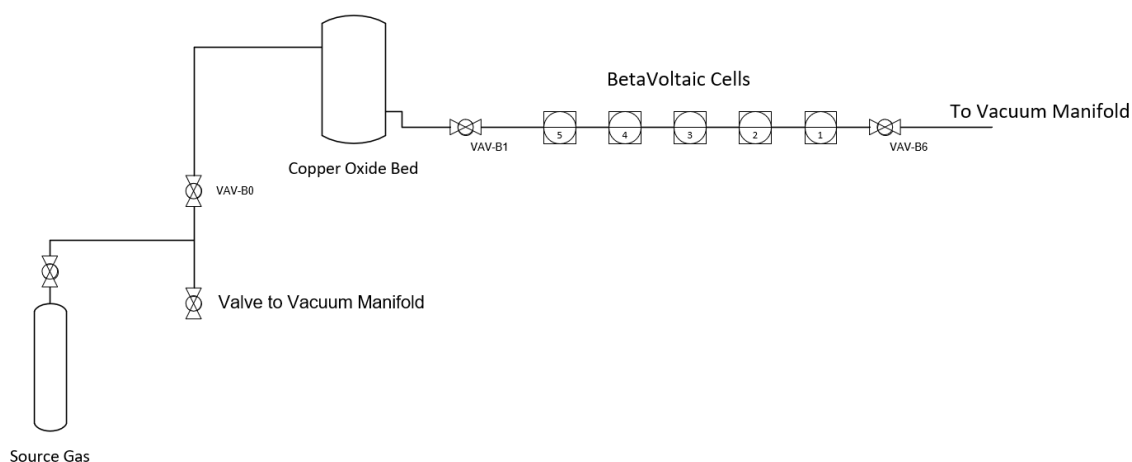


Figure 4. P&ID of the betavoltaic portion on the vacuum manifold

The final pressure inside the BVs was determined by monitoring the current output (i.e. maximizing it, final value 0.2 atm), at which time the copper gas lines were crushed and cold-welded by a manual 16-ton hydraulic crimper press and custom crimper block. The separated BVs are shown in Figure 2d.

2.6 Sensor Platforms Development

Three sensor platforms containing nW scale electronics were tested under BV power. Each platform comprised a vibrational, chemical, or optical sensor, all of whom dissipated from 5 to 375 nW of power. These sensors were chosen to exemplify the utility of such μ W scale BVs to nW electronic sensor platforms.

The vibrational sensor was a Signal Quest SQ-MIN-200 sensor mounted to the vendor development board (SQ-SEN-200-DMK) with soldered leads for power and readout. The chemical sensor was produced along the lines of (Kennedy et al., 2017) and mounted to a general development board with leads for both power and readout. Lastly, the optical sensor was an Adafruit Industries CdS photoresistor in an MCIGICM 400 points solderless breadboard, again with leads appropriately placed for readout and power.

3.0 Results

Figure 5 shows the relative power output in red with an internal pressure of 0.8 atm. The photoluminescent intensity of the absorber layer is shown in blue, normalized to that of the uncoated semiconductor. Initially, the 5 BV devices produced currents from 43-485 nA (shown in Figure 5).

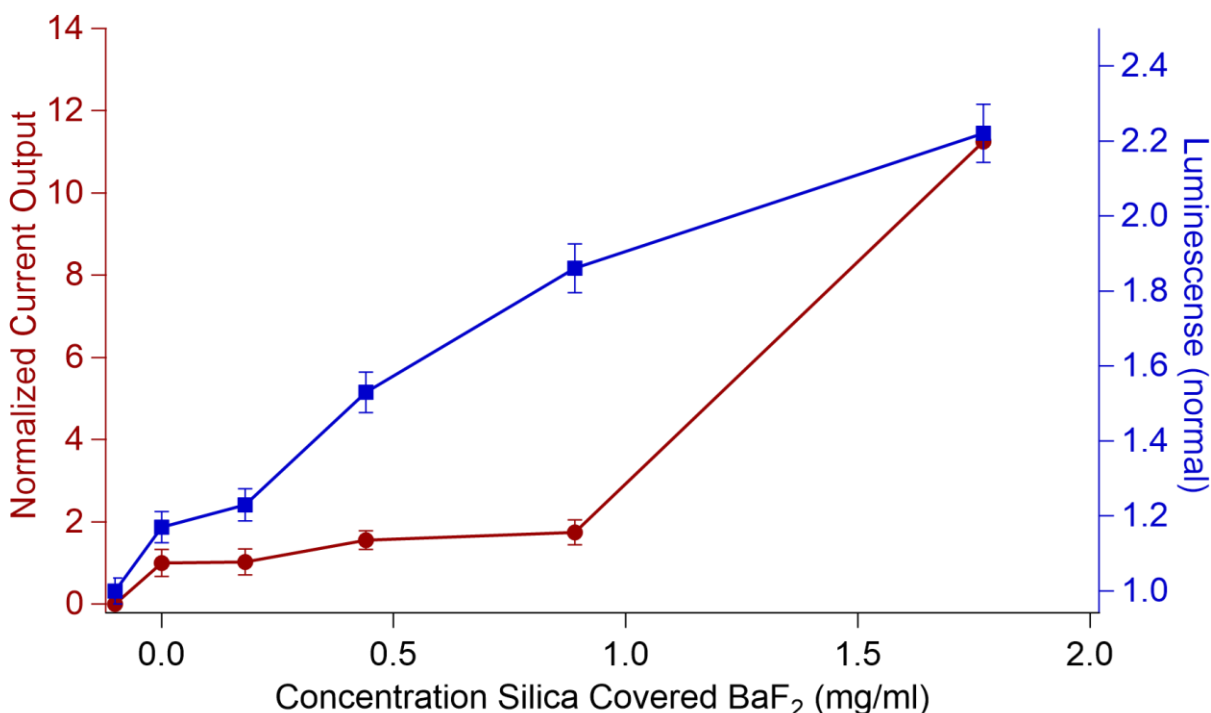


Figure 5, The normalized current output of the betavoltaics at an internal pressure of 0.8 atm and the corresponding luminescence of the tritium absorber layers as a function of the concentration of silica covered scintillator

As excess water was suspected to be shorting the devices, the internal pressure was reduced to 0.2 atm. At this time, the BV devices containing 0.18 and 1.77 mg/ml of scintillating particles began to produce $10.0 \pm 2.7 \mu\text{A}$ and $22.2 \pm 0.4 \mu\text{A}$, respectively. The currents in the other 3 devices remained relatively unchanged. The BV device producing 22 μA was selected as a power source for the sensor platforms.

The open-circuit voltage (V_{oc}) of the highest current unit was measured at $0.357 \pm 0.012 \text{ V}$ which equates to a V_{oc} of $0.071 \pm 0.002 \text{ V/chip}$, which is in line with other BV cells of this type (Alam & Pierson, 2016; Krasnov & Legotin, 2020; Svintsov et al., 2018; Zhou et al., 2021). The power of the BV 5 unit was $7.93 \mu\text{W} \pm 0.41 \mu\text{W}$. The minimum possible conversion efficiency of the device (assumes all 17.6 Ci stays in the BV getter) is $1.3 \pm 0.1 \%$. A more probable efficiency based on the absorption kinetics (assumes 10.7 Ci in the BV getter) is $2.2 \pm 0.1 \%$.

The results of the BV-powered sensor platforms are shown in Figure 6. An example readout of the vibration sensor is shown in Figure 6a. Figure 6b shows the Fourier transform of the vibration readouts of 5 similar vibration sources (1-1000 Hz ranges) that were placed next to the cart containing the sensor. Figure 6c shows the readout of an additively manufactured chemical sensor. The chemical sensor is shown to provide about 2.5 fold sensitivity between

measured gases as is in line with previous works (Kennedy et al., 2017). Lastly, Figure 6d shows the readout of the photoresistor when the ambient light is lowered by an order of magnitude. As can be seen in Figure 6, the 3 demonstrated sensor platforms pulled 5-375 nA of current when powered by the BV unit.

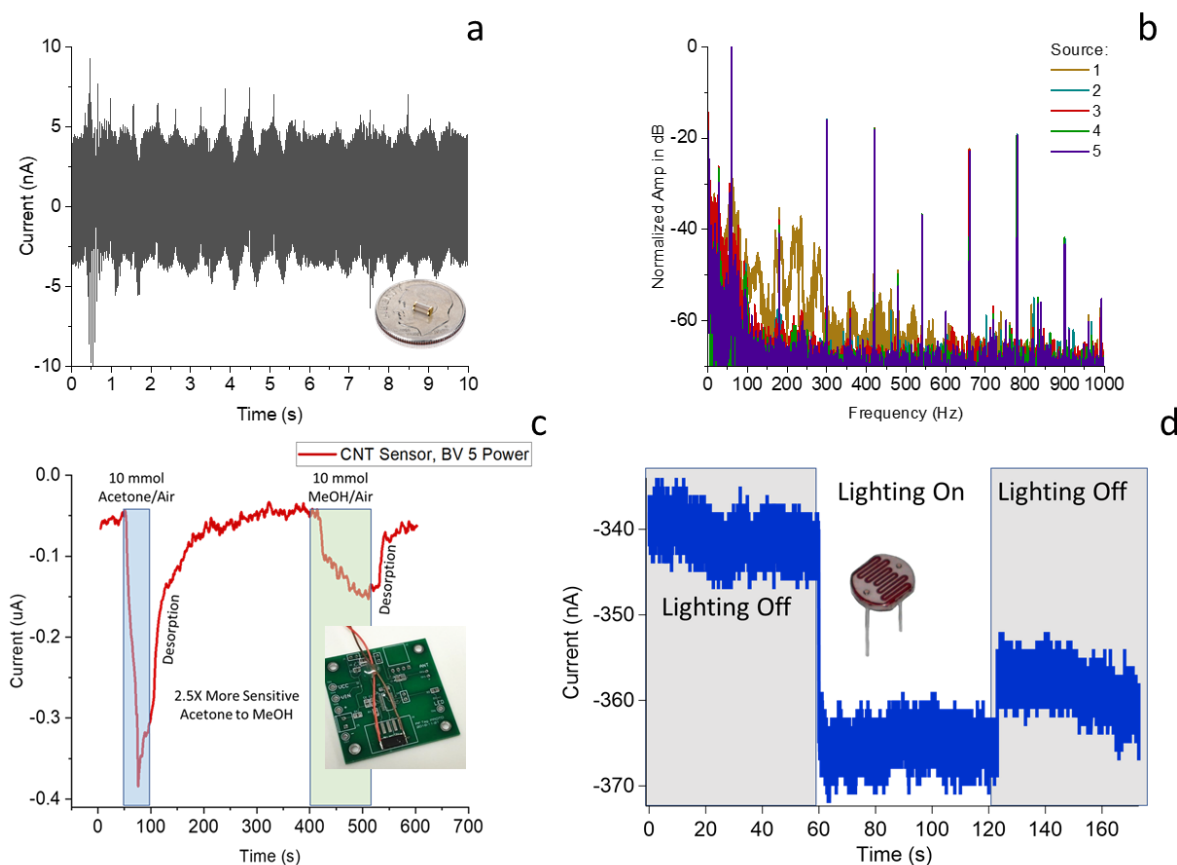


Figure 6, a) An example readout of a vibration sensor powered by a BV unit 5. B) The Fourier transform of vibration sensor data showing that the BV-sensor platform can determine the differences between 5 similar vibration sources. c) Chemical sensor data from the BV-powered chemical sensor showing a 2.5-fold sensitivity increase between acetone and methanol gases. d) Data from a photoresistor sensor-BV platform showing the readout differences as the ambient lighting level was changed from approximately 0.01 kW/m² to 0.1 kW/m² and back again.

4.0 Discussion

Current reviews of the BV design field find that laboratory grade BVs (Si p-n junction based) produce from 0.002-15.1 nW/cm² (Krasnov & Legotin, 2020; L. Olsen, 1993; Zhou et al., 2021). The control BV (BV1), which did not contain scintillating particles, produced power densities in line with current literature (0.67 nW/cm²). The addition of the scintillating particles boosted device performance significantly from 1.5 to 500 times (1-337 nW/cm²) over the concentration range of the added scintillators. These power density ranges are more typical of wider bandgap-based BVs rather than the more manufacturable Si-based units (Krasnov & Legotin, 2020; Zhou et al., 2021). While further work is needed quantifying the influence of the scintillating particles, the results presented herein support the hypothesis that the addition of scintillating particles to the getter matrix increases the device performance.

As the BV-powered vibrational platform requires 10 nA of current as seen in Figure 6a, the BV will likely continue to power the sensor for 15-80 yrs, (based on the tritium decay curve, (Traub & Jensen, 1995)). It is projected that the platform will fail when the applied voltage falls below the operational limit of the sensor. The voltage is suspected to be the main determining factor in this case, as the BV will supply the needed current for up to 80+ years.

The chemical sensor in Figure 6c required a maximum of 375 nA to power the device. As the device is a resistive sensor it will continue to provide some estimate of the chemical exposure as long as the measuring electronics can read the device. Thus, the BV sensor platform in this case will operate up to a maximum of 50-60 years, at which point the BV will not produce sufficient current to power the platform. The system lifetime will be dictated by the sensitivity of the readout electronics.

Lastly, the optical sensor platform required 375 nA to function under BV power, Figure 6d. This puts the platform at a maximum of 50-60 years of operation. As the sensor did not function well at the applied bias of the BV units (~0.3V) it is anticipated that the platform lifetime would be much shorter. As the BV 2 unit expressed a similar voltage to BV 5 (used for sensor demonstrations) and produced half the current, the optical sensor platform will likely operate for at least 15 years and up to 60 years.

The evidence of the 3 tested sensors platforms demonstrates that nW scale electronics can be powered by μ W scale BVs for several decades. Further work incorporating the scintillating particles into a more permanent getter is needed to ensure maximum lifetime of the platform, but the above work does show that it is possible for BVs integrated with scintillators in the rad source to provide usable power for decades.

5.0 Conclusions

The addition of scintillating particles to a tritiated water getter increased the performance of the silicon p-n junction-based BVs. As the two best performing BV devices produced microwatt scales of power, they are anticipated to run nW-scale electronics for several decades. Further optimization work is needed, but the incorporation of scintillating particles into BV getters was shown to boost Si-based BV performance into ranges more commonly seen for wide bandgap-based BVs. As this technique enables higher power density BV devices from conventional Si semiconductors (compared to wider bandgap devices), it is hoped that further implementation will lower the barrier-to-deployment of these long-life power platforms.

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Pacific Northwest National Laboratory

902 Battelle Boulevard
P.O. Box 999
Richland, WA 99354
1-888-375-PNNL (7665)

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