Army Research Laboratory



## Isotope Beta-Battery Approaches for Long-Lived Sensors: Technology Review

by Marc Litz

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Marc Litz Sensors and Electron Devices Directorate, ARL

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## **Executive Summary**

Long-lived energy sources are needed for remote placement of unattended electronics in sensor network applications. The energy density of isotopes exceeds that of chemical energy storage by 6 orders of magnitude. This unique energy density characteristic is offset by the fact that power output should be limited to 1 mW levels in situations involving human transport, for both radiation regulation and safety. The low-power limitation is exceeded in long-lived power for space travel applications, where 200 W isotope power sources (radio-thermal-generators) are already available for long-lived space missions (6). Isotopes are used in many commercial applications (1) and are produced and available at modest prices. As sensors get smaller, and power level requirements are reduced, the use of compact, low-power, long-lived power sources will increase. Isotope batteries are relevant in this operational regime.

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## 1. History

The first report of charge storage from isotopes was documented in 1913 (2). In the experiment, Moseley showed that in an evacuated chamber surrounding 20 mCi of radium, he could develop 100s of kV of voltage potential due to the charge emitted (beta decay) from the radium. Charge collection using isotope emission showed just how versatile were Moseley's experimental talents. It was particularly challenging at that time because few materials were available to sustain high-voltage. This result was simply described and had great impact.

Moseley was already well-known at the time for his experimental verification of the newly evolving Bohr Theory of the atom (13). Moseley's Law showed the relationship between frequency (f) of emission of characteristic x-rays from materials and their atomic number (Z).  $f(Hz) = 2.47 x 10^{15} * (Z-1)^2$  Bohr postulated that the emission of EM radiation (photons) accompanied the jumps in energy levels between the electron shells.

In the 1950s, direct energy conversion from nuclear sources was under investigation once again. Direct charge collection in vacuum for applications of long-lived voltage sources, (14) was studied by accumulating charge emitted from  $^{90}$ Sr- $^{90}$ Y for a high voltage source. Dielectric materials (15) were proposed as an intermediary for charge collection in order to avoid the need for vacuum control systems. Current generation (electron flow) in semiconductors, exposed to nuclear radiation, (16) was the next step investigated as a power source.

In the 1960s, small scale concepts for compact isotope batteries were explored further. It was not until the 1970s that the concepts were developed for medical technology implant applications, and verification testing pursued. During this time period, more than 100 papers were published in medical journals describing betavoltaic approaches for isotope-powered pacemaker replacements, with the advantage of avoiding the risks of multiple surgeries. Previously, pacemakers were powered by mercury-zinc batteries (*12*). Several companies in the U.S. (Medtronic, Cordis) (*9*) developed products. Early versions used <sup>238</sup>Pu and <sup>147</sup>Pm as the isotope source. The first successful long-term pacemaker was implanted in a 34-year-old patient in February 1960 in Uruguay by Dr. Otrestes Fiandra and Dr. Robert Rubio (*12*). In the decades that followed, fewer than 20 papers reported on the longer term success of the nuclear-battery powered pacemakers. As medical procedures became less intrusive (and safer) on the human body, the social stigma of "nuclear" power overcame the safety issue associated with fewer medical procedures.

In 2007, DARPA supported three isotope battery technologies with a goal of 35mW in 1cc volume. General Atomics (GA) led a thermo-photo-voltaic (TPV) conversion battery effort using <sup>238</sup>Pu with an 87-year half-life. RTI Inc. led a thermo-electric (TE) conversion battery effort, also using <sup>238</sup>Pu. North Dakota State University (NDSU) led a direct-conversion battery program in

boron-carbide using <sup>209</sup>Po with a 102-year half-life. Target efficiencies of 8.5%, 7.2%, and 10% were predicted. The fissioning of <sup>238</sup>Pu produces heat, which is converted to electricity in the GA and RTI programs. The direct-energy-conversion method of converting beta-emissions from <sup>209</sup>Po produced a higher conversion efficiency because the heat cycle was avoided.

Low-power applications are well-suited to using micro-grams of radiation-safe isotope. Longlived applications are particularly well-suited to isotope power sources, including long-lived power sources for sensors in remote locations, self-powered sensors on automobiles, tracking animals, buried infrastructure/building/bridge sensors, self-powered MEMS, and micro-robots. Flight recorders using <sup>60</sup>Co providing 100uW power (*11*) have been proposed for commercial use. Protections of electronics controlled by field programmable gate arrays (FPGAs) can be inhibited/destroyed for applications requiring anti-tamper (AT) circuits. The most compelling applications are those requiring energy sources for long-lived sensor elements for space, oceanographic exploration, bio-medical engineering, and structural engineering.

#### 2. Background

Isotope batteries provide a continuous flow of energy from decaying isotopes. When isotopes decay, they emit alpha, beta, or gamma particles. The particles emitted energy can be converted to electrical energy without intermediate thermalization. This is the focus of the following report. Isotopes decay in five possible modes—(1) alpha decay, (2) beta decay, (3) positron emission, (4) electron capture, and (5) isomeric transitions. Combinations of particles ( $\alpha$ ,  $\beta$ , positron, and  $\gamma$ ) are commonly emitted during a given type of decay. Alpha particles (5 MeV) typically carry more energy than  $\beta$  or  $\gamma$  emitters, but few energy conversion materials can stand-up to the  $\alpha$  bombardment over long-time periods (or short-time high flux) without deterioration.

#### 2.1 Radio-Isotope-Thermal-Generators (RTG)

Radio-thermal-generators (RTGs) have been used in many space missions because both the heat and electricity produced is useful in the space environment. Both the U.S. and Russian space programs have flown more than 50 missions, each using RTGs. Heat energy is created through the natural  $\alpha$ -decay of plutonium (<sup>238</sup>Pu). Iridium-clad, plutonium dioxide fuel (PuO<sub>2</sub>) is converted to electrical power through the use of silicon germanium (SiGe) TE cells. 300 W electrical power was generated from 4.4 kW heat using RTGs in the Galileo, Ulysses, and Cassini space missions. The 7% conversion efficiency of the heat to electrical energy is typical of TE and is sufficient for the application.

RTGs are common for space applications. RTGs produce hundreds of watts of electrical power through generating heat in surrounding local materials, then converting the heat energy to electrical energy through surrounding materials. Two paths for heat-to-electrical conversion are differentiated by the black body temperature of the heat source. TE materials use small

differences in temperature between two metals or semiconductors to create electromotive forces that generate a voltage difference. In TPV materials, the heat is converted to infrared (IR) energy, then converted to electrical in photovoltaic (PV) devices. TE is typically used below 800 °C, while TPV is more efficient above this temperature.

Nuclear energy, converted into electrical energy via the Seebeck effect, is characterized as a temperature-dependent voltage generation in materials. The nuclear energy is used to heat one side of a metal bar (or semiconducting material), causing electron with high thermal energy to flow from the hot side to the cold side, generating a DC voltage. This approach does not lend itself to miniaturization due to large thermal losses, resulting from a significant increase in the ratio of surface area to volume, upon going from a macroscale to the microscale design. NASA has been using RTGs since the 1960 in missions like Voyager and, more recently, the Cassini probe, now in orbit around Saturn.

Many RTG devices have been created for space vehicles. For this application, the heat energy is used directly for onboard equipment, in addition to the electrical power converted from heat. The conversion efficiency from heat to electrical, of these devices is ~10% (3). This order of magnitude of conversion efficiency is typical of heat-to-electrical processes (4, 5). The development over years of effort has created dozens of RTG that have been used in space missions (6). A short list of these is shown in table 1.

Spacecraft	Power Source	No. of RPSs	Mission Type	Launch Date	Location
Transit 4A	SNAP-3B7	1	Navigational	06/29/1961	Currently in orbit
Transit 4B	SNAP-3B8	1	Navigational	11/15/1961	Currently in orbit
Transit 5BN-1	SNAP-9A	1	Navigational	09/28/1963	Currently in orbit
Transit 5BN-2	SNAP-9A	1	Navigational	12/05/1963	Currently in orbit
Transit 5BN-3	SNAP-9A	1	Navigational	04/12/1964	Reentered; burned up
Nimbus B-1	SNAP-19B2	2	Meteorological	05/18/1968	Aborted; retrieved
Nimbus III	SNAP-19B3	2	Meteorological	04/14/1969	Currently in orbit
Apollo 12	SNAP-27	1	Lunar/ALSEP	11/14/1969	On lunar surface
Apollo 13	SNAP-27	1	Lunar/ALSEP	04/11/1970	Reentered in South Pacific
Apollo 14	SNAP-27	1	Lunar/ALSEP	01/31/1971	On lunar surface
Apollo 15	SNAP-27	1	Lunar/ALSEP	07/26/1971	On lunar surface
Pioneer 10	SNAP-19	4	Planetary/Sun escape	03/02/1972	Heliosheath
Apollo 16	SNAP-27	1	Lunar/ALSEP	04/16/1972	On lunar surface
Triad-01-1X	Transit-RTG	1	Navigational	09/02/1972	Currently in orbit
Apollo 17	SNAP-27	1	Lunar/ALSEP	12/07/1972	On lunar surface
Pioneer 11	SNAP-19	4	Planetary/Sun escape	04/05/1973	Heliosheath
Viking 1	SNAP-19	2	Mars Lander	08/20/1975	On martian surface
Viking 2	SNAP-19	2	Mars Lander	09/09/1975	On martian surface
LES 8, LES 9	MHW-RTG	2, 2	Communication	03/14/1976	Currently in orbit
Voyager 2	MHW-RTG	3	Planetary/Sun escape	08/20/1977	Heliosheath
Voyager 1	MHW-RTG	3	Planetary/Sun escape	09/05/1977	Heliosheath
Galileo	GPHS-RTG	2	Planetary (Jupiter)	10/18/1989	Intentionally deorbited into Jupiter
Ulysses	GPHS-RTG	1	Solar and space physics	10/06/1990	Heliocentric, polar orbit
Cassini	GPHS-RTG	3	Planetary (Saturn)	10/15/1997	Operating at Saturn
New Horizons	GPHS-RTG	1	Planetary/Sun escape	01/19/2006	En route to Pluto

Table 1. U.S. spacecraft using RTGs.

NOTE: ALSEP, Apollo Lunar Surface Experiments Package; GPHS, general purpose heat source; LES, Lincoln Experimental Satellite; MHW, Multi-hundred Watt; RTG, radioisotope thermoelectric generator; SNAP, Systems for Nuclear Auxiliary Power.

SOURCES: Data from G.L. Bennett, J.J. Lombardo, and B.J. Rock, "Development and use of nuclear power sources for space applications," *Journal of the Astronautical Sciences* 29 (October-December):321-342, 1981; N.L. Johnson, "Nuclear power supplies in orbit," *Space Policy* 2:223-233, 1986; G.L. Bennett, "Space Nuclear Power: Opening the Final Frontier," AIAA 2006-4191, p. 2, presentation at 4th International Energy Conversion Engineering Conference and Exhibit, San Diego, Calif., June 26-29, 2006.

The energy release per decay of  $^{238}$ Pu is 5593.2 keV. Smaller RTGs use as little as 7kG of  $^{238}$ Pu, which corresponds to  $1.77 \times 10^{25}$  atoms (8). With a half-life of 87.1 years, this corresponds to  $4.4 \times 10^{15}$  decays/sec (Bequerels). Therefore, energy released per second is 3973 J/s. Given the 300 W electrical output of the RTG, this corresponds to 7.5% radiation-to-electrical efficiency. Because the heat warms the electronics in the spacecraft, this heat is welcome in the emptiness of cold space. While not very efficient at generating electrical energy, it suffices for this application.

#### 2.2 Direct-Energy-Conversion (DEC)

DEC avoids the heat production process used in RTG operation. DEC can be more efficient in converting the radiation energy into electrical power, by virtue of avoiding the Carnot Cycle. The larger the change in temperature, the more efficiency can be achieved. High temperature power plants can be 58% energy efficient (7). Scaling down to smaller sizes makes achieving these efficiencies difficult. Some options for using the radiation more directly than conversion to heat are shown in figure 1. Avoiding the heat cycle can be an important step in making power sources more efficient. Each conversion step, from one form of energy to another, has loss (inefficiency) associated with it. In compact applications on earth, the highest efficiency is desirable to reduce

radioisotope activity that must be carried, and increase safety. Using the radiation decay products ( $\alpha$ ,  $\beta$ , positron, and  $\gamma$ ) directly—avoiding the heat cycle—has potential for higher efficiency power sources.

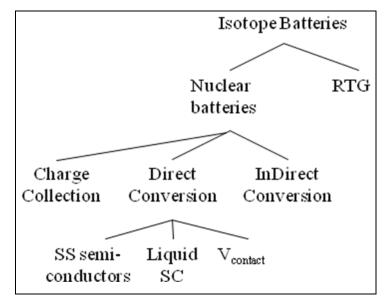


Figure 1. Decaying isotopes release  $\alpha$ ,  $\beta$ , and  $\gamma$  that can be converted to heat or create free-electrons in surrounding materials that can be collected directly or converted to create a current flow.

An early description of direct charge collection was given in 1913, creating a novel high-voltage power source (2). The charge of the emitted particles can be collected directly on a plate, which creates the power source. Transformers can also be used with reasonable efficiency to optionally transform a current source into a voltage source.

Indirect-conversion often includes phosphors that both shield the direct radiation and glow from the radiation, leading to photo-voltaic (PV) conversion to electrical energy. This two-step process includes an additional conversion process, which can reduce total efficiency into electrical current.

Direct conversion of radiation to electrical power can be performed by at least three techniques. The common thread to these approaches is the existence of an internal electric field within the physical geometry. The electric field is required in all configurations to collect and/or create additional charge carriers for electrical power output. The electric field in a semiconductor is essential in order to pull the free charges through the device, creating a flow of charge. In a chemical battery, the internal E-field can be generated in the work function differences between cathode-anode or cathode-electrolyte, or electrolyte-anode.

The radiation emitted from the isotope creates electron-hole pairs in semiconductors or electronion pairs in an electrolyte medium. The internal electric field created by the anode-cathode or doped n-p regions provides the electromotive force to move the charged particles to the oppositely charged end of the device structure. In batteries, the electrolyte (medium between anode-cathode) causes a chemical reaction in the anode and cathode (AK), creating free charges, and resulting in an electrical current between the AK. In semiconductors, electron-hole pairs (e-h pairs) are created from isotope decay product bombardment (23). Electrons in the depletion region drift creating an electrical current.

#### 2.3 Radiation Tolerance

Radiation damage in materials can limit the lifetime of use of typical semiconductors to a few years. At a minimum, it is necessary to match the reduced lifetime from beta or alpha damage to the expected lifetime of the decaying isotope. The effects of radiation damage to semiconductors include (1) increase in leakage current, (2) change in doping concentrations, and (3) shortening of carrier lifetimes. While the fluence of electrons from 1 Ci of isotope is  $3.7 \times 10^{10} \text{ e}^{-}/\text{sec}$ —low compared to that of accelerators—the long lifetime expectation of power sources creates total radiation dose that can become significant over the lifetime of the device and must be considered in end-of-life design.

The basic mechanisms for radiation damage in semiconductor devices are displacement damage, which alters the crystal structure, and ionizing damage liberating charge carriers altering expected device operation. Displacement damage depends on mass and momentum of incoming particle and the interaction with the lattice (non-ionizing energy loss). Ionization effects depend more on energy absorbed rather than radiation type. The energy absorbed is measured and characterized by dose (1 Gy=1 J/kg). Mitigation techniques against damage to materials will vary depending on semiconductor material, particle type, particle energy, and device geometry.

Wide band-gap (WBG) materials (i.e., SiC, GaN, AlN, Diamond) that exhibit tight interwoven crystal structure, small lattice constant (atomic spacing), and high quality material (defect free) are a few of the characteristics that effect radiation tolerance. Of the WBG materials listed, SiC is the most mature in manufacturing and device geometry design capability (24). The mismatch of lattice constant between GaN and the epitaxial growth layer (sapphire) causes dislocations in devices. This is not a problem in LED applications. Continued development may help overcome this issue. The largest ionic bond strength and material density (6.2 g/cc) make GaN a material of interest for the future.

Radiation damage is highly dependent on, and, therefore, a strong function of, bombarding particle, particle energy, and semiconductor material. It is difficult to make direct linear comparisons across these characteristics. Material properties, stopping range of charged particles, and particle mass are not linear in materials. Criteria for comparing materials across these boundaries are still under investigation; however, many correlations within these categories have been described (25). Increasing particle energy generally reduces the lifetime of a given material. Large band gap materials tend to increase the radiation tolerance of an energy converter, providing higher efficiency conversion for longer periods of time.

One very interesting approach towards minimizing radiation damage is through the use of liquid semiconductors. They exist at high temperatures, and are being investigated for their increased radiation tolerance because of the self-annealing properties of the liquid-state (22).

### 3. Beta Voltaic Approaches

The use of beta (electron)-emitting isotopes is most common in isotope batteries. Low-energy (<250 kV) electrons are stopped by thin metallic foils (20). This characteristic helps significantly with any safety concerns by collecting unconverted energy in a metal foil shielding. The low energy requirement on the emitted beta also limits possible production of bremsstrahlung radiation. The electrons can be collected in an absorber or converter. If the absorbing material is the depletion region of a SiC semiconductor, for example, then electrical current is produced at the voltage proportional to the band gap (21). The higher energy electrons are usually slowed down in the material, creating many knock-on electrons along the way in the material. The lower energy (~1 keV) knock-ons are collected and swept out in a depletion region of a semiconductor. The electrons can then be used directly to create a circuit current. Output voltage can be increased by cascading several junctions in series. Output current can be increased by adding junctions in parallel or increasing junction area.

#### 3.1 Direct Energy Conversion

All that is required for the electrons to be swept out is an internal field. The sandwich of p-n junctions surrounding the beta emitter is shown in figure 2. The betas emitted from the isotope layer create electron-hole pairs in the semiconductor. The internal electric field in the depletion region (in between the p-n layers) pulls the electrons in and out, creating a trickle charge of current. Each p-n junction will produce a voltage related to its built-in field and internal resistance.

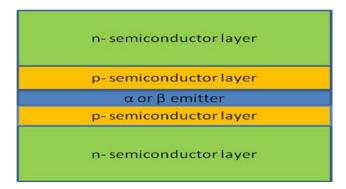


Figure 2. The most straightforward approach is to utilize direct-energy-conversion. Alphas, betas, or gammas create electron hole pairs in semiconductors. The internal electric fields of the semiconductor pull out the free-charge and current flow is generated.

Semiconductors materials with higher band gaps, such as SiC, GaN, and diamond, are more radiation-tolerant than Si. Si is the least expensive semiconductor material. The bandgap correlation to radiation tolerance is a first indicator.

#### 3.2 Indirect Energy Conversion

The indirect approach described here is a two-step process. Betas emitted from the isotope are collected in a phosphor. The phosphor glows, creating optical photons. The optical photons are then collected in a standard photo-voltaic (PV) cell, creating electrical current at the voltage of the PV cell. The two-step process, shown in figure 3, has the value that the betas can be shielded in the phosphor before getting to the semiconductor layer. Secondly, the PV converter is a more commercially available device that can be less expensive than the design and optimization of direct-energy-converter semiconductors.

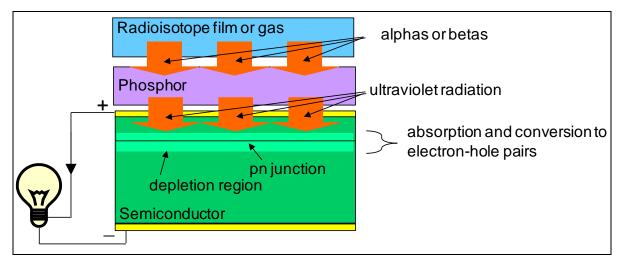


Figure 3. The two-step process of indirect energy conversion is shown. Radiation from decaying isotopes creates excitations in a phosphor which emits optical photons. The optical photons are collected by photovoltaic cells and converted to electrical current.

The disadvantages of this two-stage configuration are that it may be less efficient, as there are two steps to convert to electrical power, with associated losses. Each energy conversion step has its own efficiency. It can suffer from being less compact than a direct process, and it is a more complex structure to fabricate. The advantage that it offers is that there is less direct degradation of the lifetime of the Si or GaAs PV semiconductor, as there is little or no direct beta bombardment of the semiconductor material.

## 3.3 Hybrid Approach

A design based on both direct-energy conversion (DEC) and indirect energy-conversion has been developed in order to maximize the power output of the device. The concept is illustrated in figure 4. Mixing the decaying isotope of <sup>155</sup>Eu in with a gadolinium flouride phosphor is a novel approach that is predicted to be more efficient at generating optical photons, as the mixture is

made homogeneous (18). Another novel aspect to the approach is that the phosphor is doped with europium (GdF<sub>3</sub>:Eu). The Eu doping gives it a characteristic optical output of 620 nm, which matches well with the common photovoltaic (PV) response function.

It is hoped that this system can be optimized for maximum power output and long-lifetime. The ratio of  $^{155}$ Eu to GdF<sub>3</sub> has been varied to minimize optical attenuation, and the homogeneous mixture enhances phosphor conversion into optical photons. The thickness of the GdF<sub>3</sub>:Eu had been optimized for minimum attenuation of the optical photons by the medium, yet gathering the remaining low energy betas for energy harvesting, as well. By matching the beta energy of the  $^{155}$ Eu to the thickness of the phosphor layer, the search for the maximum power point is underway (*18*).

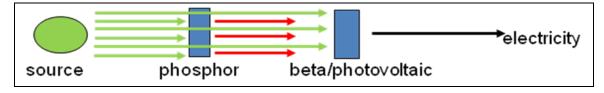


Figure 4. Hybrid geometry with GdF<sub>3</sub> phosphor uses both direct and indirect conversion methods to offer best lifetime and power performance.

## 4. Design Concept using Commercially Off-The-Shelf Components

The characteristics desired for unattended sensor are long-life operations and safety. There are several beta emitters that exhibit long-life. Long-life, in this case, means the lifetime of infrastructure, ~150 years. Keeping safety first means (a) limiting gamma emission and (b) keeping the amount of radioactive material as low as is reasonably possible, while still providing 100  $\mu$ W trickle charge output.

Technology issues associated with developing isotope batteries include choices of isotope material, approach to converting energy stored in nucleus to electrical power, and power management. Non-technical issues include packaging of the device and licensing of the packaged system. The technical issues are more interesting and definable; however, the non-technical issues have limited the development of useful devices.

The radioisotopes (RI) shown in table 2 are primarily beta emitters. The most compelling interest in a RI power source is the unique niche of long-lived power unmatched in chemical power sources. The potential for nano-sized power sources exists because of the isotope energy density; however, energy conversion in the balance of the system presently accounts for a much larger volume in this type of device. The list of isotopes with lifetimes longer than 10 years and beta energies lower than 250 keV are each highlighted in the table. Of the reduced list, 63Ni and 3H are the most readily available, as they are used in commercial applications. One of the highlighted isotopes exists in an isomeric form, <sup>121</sup>Sn, which is of interest in the future for the capability to be triggered, offering the possibility of long-shelf-life, before releasing the energy stored.

						beta	
Α		Ζ	N	halflife	alpha	pk kev	IT,ec
3	H(T)	1	2	12.32		18.6	
14	C(PMMA)	6	8	5710		156	
32	P	15	17	0.04		1711	
35	S	16	19	0.239		167.4	
42	Ar	18	24	32.9		600	
44	Ti	22	22	49.3		266	
55	Fe	26	29	2.73		232	
60	Co	27	33	5.24		310	1170,1330
63	Ni	28	35	101		66.9	
85	Kr	36	49	10.755		687	
90	Sr	38	52	28.77		546	
106	Ru	44	62	1.0234		39	
109	Cd	48	61	1.2674		184	
113	Cd	48	65	14.1		58	
121	Snm	50	71	55		6	
137	Cs	55	82	30		190	
145	Ρm	61	84	17.7		161	
147	Ρm	61	86	2.624		225	
108	Agm	47	61	418.25			109.5,2027
108	Agg	47	61	4.50913E-06		1649	
148	Gd	64	84	75	3271		
151	Sm	62	89	90		76	
155	Eu	63	92	4.67		253	
157	Tb	65	92	99		63	
171	Tm	69	102	1.92		96	
178	Hfm	72	106	31			2445
179	Ta	73	106	1.79		110	
198	Au	79	119	8.56164E-08		980	
193	Pt	78	115	50		56	
204	TI	81	123	3.78		763	
210	Рb	82	128	22.29		63	
208	Po	84	124	2.8979	5216		
210	Po	84	126	0.379	5304		
228	Ra	88	140	5.75		46	
227	Ac	89	138	21.773		44	
228	Th	90	138	1.9131	5520	Beta emitte	rowith
232	U	92	140	68.9	5414		
235	Np	93	142	1.085	123	$\tau_{\gamma_2}$ >12 yrs a	
236	Pu	94	142	2.857	5867	Ewn<250 ke	٧
238	Pu	94	144	87.74	5593		
241	Pu	94	147	14.35		21	
241	Am	95	146	432	5500		
242	Am	95	147	0.001826484		645	
242	Am m	95	147	141			49
243	Am	95	148	7370	5280		
244	Cm	96	148	18.1	5902		
248	Bk	97	151	9	5793		
250	Cf	98	152	13.07	6128		

Table 2. Isotopes documented in power source literature.

A short list of beta emitters that live 90–150 years is shown in table 3. From this list we can see that there are several long-lived isotope candidates. These isotopes are low gamma emitters, in

addition to having long half-lives. They are not commonly produced for industry, making their cost higher. The cost can be lowered if the commercial use increases; as more material is required the process can be made efficient.

RI	HalfLife (yr)	Eavg (keV)	Ci
<sup>63</sup> Ni	101	17	10
<sup>157</sup> Tb	71	16	2.5
<sup>151</sup> Sm	90	25.3	7
$^{3}H$	12.5	6	27
<sup>155</sup> Eu	4.67	84.3	2
<sup>147</sup> Pm	2.6	75	2.5
<sup>241</sup> Am	432	5200	0.1

Table 3. Lifetime and power output of beta emitting RI is shown. The lifetime of 90 years enables new capabilities in sensor and microelectronic power applications. The activity (Ci) is shown for 100  $\mu$ W power output assuming 10% conversion efficiency.

Sealed radiation sources are one of the important tools available in affecting safety and handling. Sealed Source is a term used to describe radioactive sources that have been designed to prevent spread of radioactive material under normal working conditions. Here is the definition from the State of Oregon Rules for Control of Radiation:

"Sealed Source' means radioactive material that is permanently bonded or fixed in a capsule or matrix designed to prevent release and dispersal of the radioactive material under the most severe conditions which are likely to be encountered in normal use and handling.

#### (Oregon Administrative Rules (OAR) 333-100-0005)

Examples of sealed sources include

- Nickel-63 Electron Capture Detector (Ni-63 ECD)–the radioactive isotope Ni-63 is electroplated to a metal foil. The foil is installed by the manufacturer in an inaccessible chamber inside the ECD cell.
- Cesium-137 (Cs-137) or Cobalt-60 (Co-60) gamma sources for radiography. These usually contain a small, pea-sized source that is sealed in a small welded capsule. The capsule is encased in a shield, usually lead, with a small shuttered opening that controls the gamma beam.

Micro-encapsulation of radiation sources reduces the possibility of leakage and makes geometries other than 2D an option for more complete interaction of radiation with converter. Shock-absorbing encapsulation adds another layer of protection supporting shipping and transportation safety. A welded metal outershell creates yet another layer of structure against shock, vibration, and leakage. The package format (e.g., size, shape) makes the power source usable for specific applications. User requirements shape the packaging—whether configured in

a cylindrical package of D-cell type for fit into electronics, or packaged in a rectangular BA5590 geometry to be a replacement for military communications power source.

Licensing with the Nuclear Regulatory Commission (NRC) eventually comes into play as the technology develops. Licensing cannot wait until the fielding of a system and is best addressed while the design is being developed in the laboratory. In this way, a redesign of the package later in the development can be avoided. The NRC performs engineering and radiation safety evaluations of the ability of sealed sources to safely contain radioactivity under transportation and industrial conditions. These evaluations are summarized in registrations that NRC maintains in the National Sealed Source and Device Registry (NSSDR). The results of these evaluations, and detailed information on the source design, function, and limitations on use, are also contained in the NSSDR. Transportation is made easier and broader use of power source can be applied if a special form qualification can be acquired through representative solid radioactive capsule evaluations. Qualification of special form for radioactive material is addresses in NRC Regulation 10CFR part 71.75. A general license can be issued by the NRC to transport, or deliver to a carrier for transport, licensed material in a package for which a license or certificate of compliance (CoC) approval has been issued.

The discussion or aforementioned process indicates the extraordinary regulation surrounding isotope-based power sources. The approach described in the next section is motivated by the overwhelming effort required to license a power source design and subsequent device.

#### 4.1 Tritium PV Approach

A short list of beta emitters that live 90–150 years is shown in table 3. From this list we can see that there are several long-lived isotope candidates. These isotopes are low gamma emitters, and also have long half-lives. They are not commonly produced for industry, making their cost higher. The cost can be lowered if the usage increased.

#### 4.2 A Commercial-Off-The-Shelf (COTS) Proposal: <sup>3</sup>H-Green Phosphor-PV

Tritium encapsulated in glass vials, lined with phosphor, has met all the levels of examination that have earned a general license for commercial lighting systems. These devices are used in emergency exit signs in airplanes and public buildings. Extending their use to provide power as a long-lived battery is a step that should be achievable in a short period of time. The commercial companies that package the light sources for exit signs and emergency lighting can provide the cells, which we propose to use as a light emitter for a power source. By adding photovoltaic conversion of the light to electrical current, a small ( $100 \mu$ W) power source can be developed. The device would be inexpensive compared to other approaches that offer more efficient conversion techniques, higher energy-density, and more compact packaging. While iterative steps can be taken to make a tritium power source more efficient (enscapsulation techniques, enhanced packaging, reduced phosphor grain size), these rudimentary sources can be useful. The impact of long-lived power sources can begin to have impact on sensors and network arrays now.

Each rectangular tritium plate holds 100 mCi tritium. Two plates are contained within a unit cell, shown in figure 5. In this configuration there are 113 mCi/cm<sup>2</sup>. The beta-energy from tritium has an end-point of 18 keV, though the average beta energy is ~6 keV. The measured beta spectrum for tritium is shown in figure 6. The nuclear power ( $W_{nuc}$  from beta particles) emitted from one tritium light source panel (see figure 5) is then ( $113x10^{-3}$  Ci/cm<sup>2</sup>)( $3.7x10^{10}$  dec/Ci/s)(6000 eV/dec)( $1.6x10^{-19}$ J/eV)=4.01  $\mu$ W/cm<sup>2</sup>. This is the nuclear power ( $P_{nuclear}$ ) emitted per square cm, from one side of the rectangular plate.

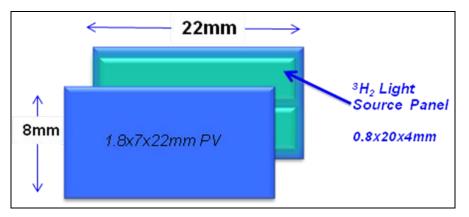


Figure 5. The unit cell consists of two tritium-phosphor platelets, surrounded by solar cells.

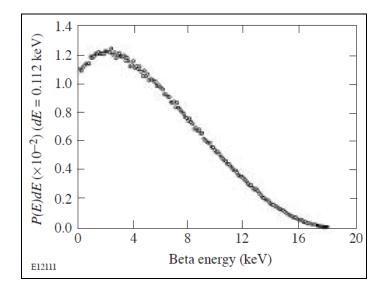


Figure 6. Energy spectrum of beta particles emitted from tritium (28).

How much activity is required to generate  $100 \ \mu W_e$ ? If we assume that commercially available photovoltaics (PV) are ~20% efficient, then we need 500  $\mu W_{opt}$  out of the PV. In order to generate this 500  $\mu W_{opt}$ , we must determine the efficiency of the phosphor in converting nuclear power into optical power.

The efficiency of the tritium-phosphor (see mbMicrotec spectral curve data sheet) is  $\sim 8.24 \times 10^{10}$  photons/cm<sup>2</sup>/s for the standard 25 mCi source. The measured spectral output of the phosphor is

shown in figure 7. A green photon is 528 nm ( $5.68 \times 10^{14}$  Hz, 2.35 eV) or  $3.76 \times 10^{-19}$  J. The manufacturer can load the tritium plate with as much as 100 mCi of tritium gas. This is limited by the tritium gas pressure in the plate. The resulting optical power from one side of the plate shown in figure 5 is ( $8.24 \times 10^{10}$  ph/cm<sup>2</sup>/s)(4)( $3.76 \times 10^{-19}$  J/ph)=124nW<sub>opt</sub>/cm<sup>2</sup>.

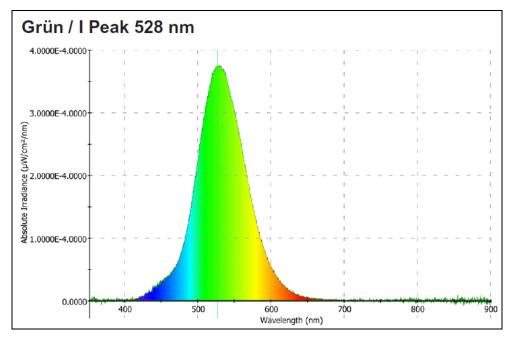


Figure 7. Spectral output of ZnS:Cu phosphor used by mbMicrotec in lighting markers.

From this optical power, we can calculate that 248 cm<sup>2</sup> of tritium-loaded plates are required to generate 500  $\mu$ W<sub>opt</sub>. Because we can couple optical power from both sides of the plate, we only require 124 cm<sup>2</sup> of tritium plates. Each plate is 0.88 cm<sup>2</sup>; therefore, 141 plates at ~\$6 each are required to complete the 100  $\mu$ W<sub>e</sub> energy source.

The ratio of optical power generated from nuclear emissions per cm<sup>2</sup> ( $P_{opt}/P_{nuc}$ ) is calculated to have an efficiency of 3.1%. The ratio of electrical power from optical power ( $P_e/P_{opt}$ ) is expected to be ~20%, depending on the PV available at low illumination. The total efficiency of the whole device, consequently, will be less than 1%. The low efficiency can be overcome in the future by using more optimized phosphors and specially made PV (where device cost will increase). We propose to show the value of extended life power sources at low cost for a first step, before proceeding to a more optimized 10% efficiency version with higher cost.

Concepts for isotope power sources over the years have included a large array of radioisotope possibilities. The RI shown in table 2 are primarily beta emitters. The primary interest in an RI power source is the unique niche of long-lived power unmatched in chemical power sources. The list of isotopes with both lifetimes longer than 10 years and beta energies lower than 250 keV is highlighted in the table. Of the reduced list, <sup>63</sup>Ni and <sup>3</sup>H are the most readily available, as they are used in commercial applications. One of the highlighted isotopes exists in an isomeric form,

<sup>121</sup>Sn, which is of interest in the future for the capability to be triggered, offering the possibility of long-shelf-life, before releasing the energy stored.

The most straightforward approach to getting a power source out in the field for trials is described previously. The approach utilizes components that are commercially available. This completely commercial-off-the-shelf approach represents an inexpensive route to producing and licensing a long-lived power source quickly. The device is not the most efficient, most energy dense, or novel in design. It is simple and straightforward, with the goal of getting this extended lifetime power source into the hands of users. We believe that this step is essential in developing the new long-lived applications that will be enabled with these power sources. Because of the extended lifetime capability, these types of power sources can have great impact towards persistent sensing, communications signal relay, and unattended remote sensing.

## 5. Safety, Availability, Common Applications

Isotope decay has superior energy density in the nuclear energy source. Minimizing the isotope activity to ultra-safe levels reduces the power output required for many applications. The power can be scaled up to meet more power hungry applications, but this requires large quantities of isotopes. High atomic number materials, like lead, are required to shield gamma radiation. Most electron energies can be stopped by a few centimeters of plastic. Alphas are stopped within  $50 \,\mu\text{m}$  of almost any material.

The concern over radioisotope (RI) materials in our society is greater than the actual health hazard. We understand the dose deposition profiles for radioactive materials in skin, tissue, and many materials. The possibility of using isotope batteries depends on offering technical capabilities that exceed the real and perceived safety hazards. The fear exists that proliferation of materials can lead to dirty bomb threats. The amounts required for isotope batteries are small compared to other more commonly found RI materials in commercial applications (7). This would force terrorists to work very hard to accumulate sufficient activity levels to become an active threat.

Radio-luminescent "EXIT" signs contain radioactive tritium (20 Curies). They are often found in airports, airplanes, schools, and hospitals, because they reduce risk of accidents in the event of power outages. Medical applications of gamma-emitting <sup>60</sup>Co include cancer therapies and bacteria sterilization. Agricultural sterilization of silkworms calls for 5 kCi of <sup>137</sup>Cs. Cesium-137 has been employed in a variety of industrial measurement gauges, including moisture, density, leveling, and thickness gauges.

The safety issues specific to power generation are small compared to previously described medical and safety signage.  $^{63}$ Ni emits low-energy betas that are stopped within 50  $\mu$ m of semiconductor material. The short penetration depth correlates to little or no-shielding

requirement. A 10  $\mu$ W power source can be built using 1 Curie ( $3.7 \times 10^{10}$  Bq). The number of curies required to fabricate a 100  $\mu$ W isotope battery are shown in table 3. The electrical power output of a battery composed of these isotopes assumes a 10% efficiency in converting from beta particle energy to electrical current in a load. The lifetime of 90–100 years for <sup>151</sup>Sm and <sup>63</sup>Ni provides a new level of capability to remotely located sensor applications. These materials are chosen because of their nearly non-existent level of gamma radiation in the beta decay.

Isotopes are commonly used in medical diagnostics and therapies. The suppliers for medical isotopes are different from those supplying isotopes for industrial use described below. Medical suppliers include Covidien, Chalk River (Canada), and Missouri University Research Reactor (MURR). Other national reactors in Belgium, France, and the Netherlands contribute to the daily requirements for short-lived medical isotopes.

Examples of radioactive tracer medical procedures include <sup>99</sup>Tc heart blood flow mapping, <sup>99</sup>Tc bone infection diagnostic, <sup>18</sup>F positron emitter for general positron emission tomography (PET), <sup>64</sup>Cu for PET imaging in tumors, and <sup>125</sup>I for prostate and brain diagnostics. The list of diagnostic medical uses is long (*17*). A few of the surgical and therapeutic applications, reduced from an equally long list, include <sup>60</sup>Co gamma knife surgery for tumor specific placement; <sup>89</sup>Sr, <sup>186</sup>Re, and <sup>153</sup>Sm to relieve cancer-induced bone pain; and <sup>131</sup>I for treating kidney, prostate, and urinary tract dysfunction and obstructions.

Isotopes are available commercially from several companies. QSA Global supplies <sup>63</sup>Ni, <sup>241</sup>Am, <sup>60</sup>Co, and <sup>252</sup>Ca for industrial applications. The Eckert and Zeigler Isotope Products Laboratory sells a large variety of isotopes for calibration systems, medical imaging, and industrial measurement. NRD Inc. supplies <sup>210</sup>Po encapsulated in Ag/Au foils for industrial de-ionizing applications. These materials are used during the fabrication of components for thickness quality control, charge neutralization, materials analysis, and sterilization. Commercially available isotopes and applications are shown in figure 8.

Extensive use of short-lived isotopes in the medical community has been discussed. As one of the larger commercial suppliers of radioisotopes, QSA sells a large variety of the commonly used isotopes to hospitals, but also offers many longer-lived isotopes for environmental monitoring applications. These include <sup>14</sup>C for dust monitor in the air, <sup>252</sup>Cf for mineral analysis, and <sup>63</sup>NI for gas sniffing in environmental quality control.

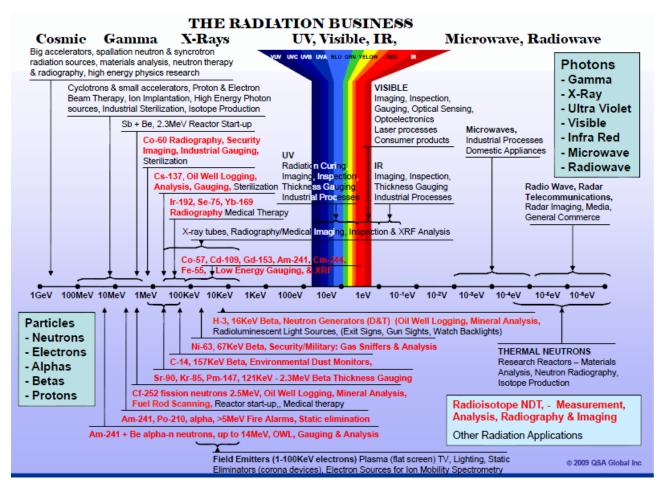


Figure 8. The commercial production of isotopes for both scientific and commercial applications is shown from the point of view of a global supplier of isotope sources. Materials analysis for security applications has surpassed that of scientific applications.

#### 6. Conclusion

The size of electrical devices is continually shrinking. The power requirements of many sensors and communications equipment can greatly reduce the power requirements of many devices, such as sensors, light sources, and transmitters. Chemical batteries are the mainstay of power for these devices. However, chemical batteries have limited lifetimes. This makes remote use and replacement difficult for applications extending the lifetime use. The most compelling reason to use isotope power sources for remotely located unattended sensors and communications nodes is the long lifetime power source capability.

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## **Appendix A. Circuit Model**

A standard PN junction geometry, illustrated in figure A-1, is sufficient to describe the direct energy conversion process of radioisotope beta emission into electrical current. The maximum current that can be generated in the direct energy conversion device is proportional to the charge of an electron(C), beta flux  $\varphi$  (#/s), and the incoming beta energy divided by the energy required to create an electron-hole pair,  $\varepsilon$  (~10 eV). The internal electric field of the semiconductor then pulls the low-energy electron created out of the depletion region, contributing to electrical power output. This process has other losses associated with it, lattice vibration phonons, energetic electrons, incomplete loss in the beta, etc. The efficiency for conversion is dependent on the energy of the beta, the semiconductor material, and crystal structure, and is macroscopically described with an efficiency constant  $\eta$ .

The energetic particles emitted during the decay of the radioisotopes create a multitude of electron-hole pairs within the semiconductor. Once the carriers are generated, positive charges flow into the semiconductor and lead to a generated current Ig, which is equivalent to a short circuit current (Isc). The displaced carriers effectively forward bias the p-layer, hence creating a voltage potential across the device. The forward bias junction yields a current (Id) through the junction that is opposite to the generated current. In the presence of very small load (short), Isc = Id, while the output voltage with very high load (open) is Voc, the open circuit voltage. When a load is connected to the diode's output terminals, as in figure A-2 the current due to the forward bias junction is split between the current into load (Iout) and Id, such that Ig = Id + Iout.

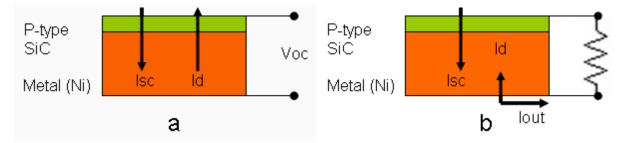


Figure A-1. Current flow under (a) open circuit and (b) load conditions.

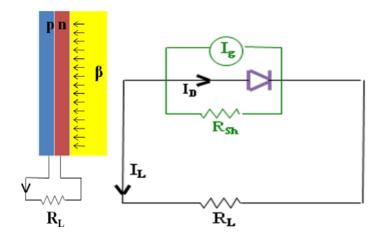


Figure A-2. Circuit diagram for a p-n junction conversion of radiation induced e-h pairs into a trickle charge of electrical current.

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