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MCNP simulation of mixed isotopes Betavoltaic Nuclear Batteries

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1. Introduction

New energy sources are being constantly developed and improved. Nuclear batteries fit in this scenario, being a technology that produces electricity through radioactive decay. In circumstances where maintenance is difficult or impossible, like as space exploration, oil production, or even in a heart pacemaker, the goal of these systems is to deliver dependable energy [1]. There are many nuclear batteries types. Among those, betavoltaic nuclear batteries (BNB) are of particular interest due to the prospects for use in microelectromechanical systems (MEMS). New generation MEMS and semiconductor devices require short-term power supplies and long operating times [2]. Often these devices are installed in places where maintenance is difficult or when data is necessary even with a power shortage [3-5].

Paul Rappaport proposed using semiconductor materials to generate electricity from beta decay energy in 1953. Atoms in a semiconductor are ionized by beta particles released by a radioactive source, resulting in decompensated charge carriers. Charges move in one direction when a p-n structure's electric field is present, producing an electric current [6].

Nuclear batteries use radioactive isotopes with half-lives that range from a couple to hundreds of years. Unfortunately, the technology has contradicting characteristics. The shorter the half-life, higher the emitted power will be. The longer the half-life is, longer the shelf life will also be [7]. Also, betavoltaic cells' power density is far lower than that of their galvanic counterparts. Despite this, betavoltaic started being employed in low-load electronic circuits in the 1970s. [6].

Various betavoltaic microbattery models based on various semiconductor materials and radioactive sources are currently being researched. The majority of the radioisotopes used in this field are challenging to manufacture, purify, and only a small number have the features required for their usage [8].

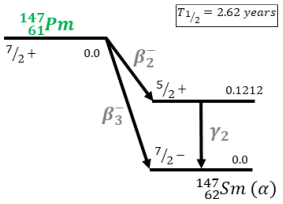
So far, mostly Pm^{147}, Ni^{63}, and H^{3} were used for fabricating BNBs. They were, up to now, proposed as a single isotope source. Mixing isotopes might allow the end battery to have a higher shelf-life and power.

The objective of this work is to create an MCNP® routine to calculate particle number of mix isotope battery and compared with the un-mixed version. Combining the isotopes strengths may minimize their disadvantages and create a better product.

2. Materials

The isotopes used for comparison will be presented next.

Pm^{147} is an ideal candidate for use in nuclear batteries due to its favorable power density (340.37 mW/g from Pm_2O_3) and low biological hazard as a result of the emission of very few low-energy gamma photons [9]. Pm^{147} is mainly produced by nuclear activation of highly enriched targets of the parent nucleus Nd^{146} or as a product of nuclear reprocessing. Figure 1 show a summary of its characteristics.



Energy informations (keV)	
β_{max}	224.60
$\beta_{average}$	66.93
γ	121.22 (0.003%)
X-rays	40.11 (0.001)

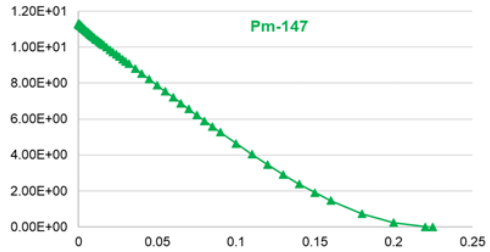
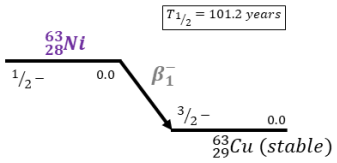


Fig.1: Pm^{147} energy emission spectrum and information

Ni^{63} is produced artificially in a high flux isotope production nuclear reactor through the (n,γ) reaction using enriched Ni^{62} or natural Ni as target [10]. Mostly enriched targets are used to minimize the need for purification. It has a low power density, 5.80 mW/g, but it has a long half-life, thus a higher shelf-life. It has even lower biological hazard (no gamma photons). Figure 2 show a summary of its characteristics.



Energy informations (keV)	
β_{max}	66.94
$\beta_{average}$	17.42

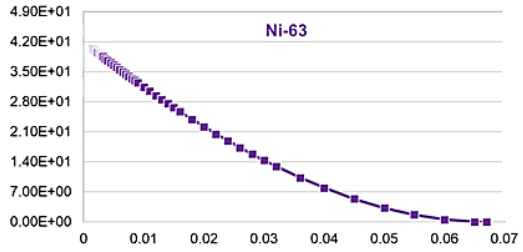
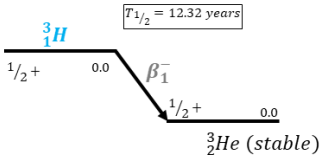


Fig. 2: Ni^{63} energy emission spectrum and information

Tritium can be recovered from reactor water and then reacted to a molecule for source fabrication. During the operation of pressurized water reactors (PWR's) a significant amount of tritiated water is continuously generated and extracted. H^3 has a high power density, 324.91 mW/g, but it has an even longer half-life than Ni^{63}. It also has lower biological hazard (no gamma photons) [11, 12]. Figure 3 show a summary of its characteristics.



Energy informations (keV)	
β_{max}	18.59
$\beta_{average}$	5.68

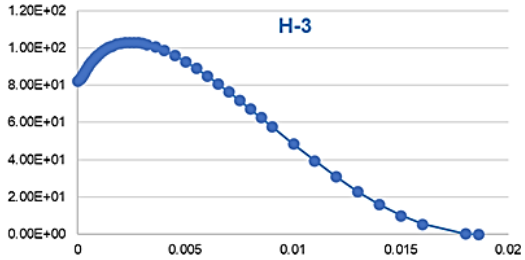


Fig. 3: H^3 energy emission spectrum and information.

3. Methodology

The radiation quantification was performed by MCNP6® Monte Carlo Code. MCNP6® is a general-purpose, continuous-energy, generalized- geometry, time-dependent, Monte Carlo radiation-transport code designed to track many particle types over broad ranges of energies. The code was developed by Los Alamos National Laboratory, USA. Figure 4 show the geometries used in the program imput.

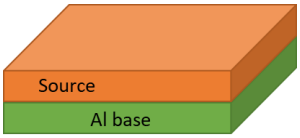


Fig. 4: Imput geometry. Surface area is 10x10 cm, the backing aluminum base has 0.01 cm. Source thickness varied.

Tally f2 (flow of particles through a surface) was used to calculate the number of particles flowing through the source top surface area. The beta decay spectrums (presented in figs. 1-3) were obtained in ICRP Decdata program. Radioactivity used varied in percentage for each isotope. Different thickness values were used respecting the thickness self-absorption cut-off.

Table 1 shows the composition of the materials used and density.

Table 1: Material compositions used in MCNP6 simulations

Material	Atomic composition	Density (g/cm³)	Reference
Backing material	Al 100%	2.70	[13]
$^{147}Pm_2O_3$	Pm^{147} 100% O 100%	6.61	[14]
^{63}Ni	Ni^{63} 100%	8.9	[15]
$^{3}H_2O$	H^3 100% O 100%	1.21	[16]
Air (wgt fraction)	C 0.000124 N 0.755268 O 0.231781 Ar 0.012827	0.001205	[17]

4. Results

3.1 Flow of particles

a) Figure 5 shows the results for Pm^{147} and H^3 Battery mix;
b) Figure 6 shows the results for Ni^{63} and H^3 Battery mix;
c) Figure 7 shows the results for Ni^{63} and Pm^{147} Battery mix;

*BOO: Beginning of Operation

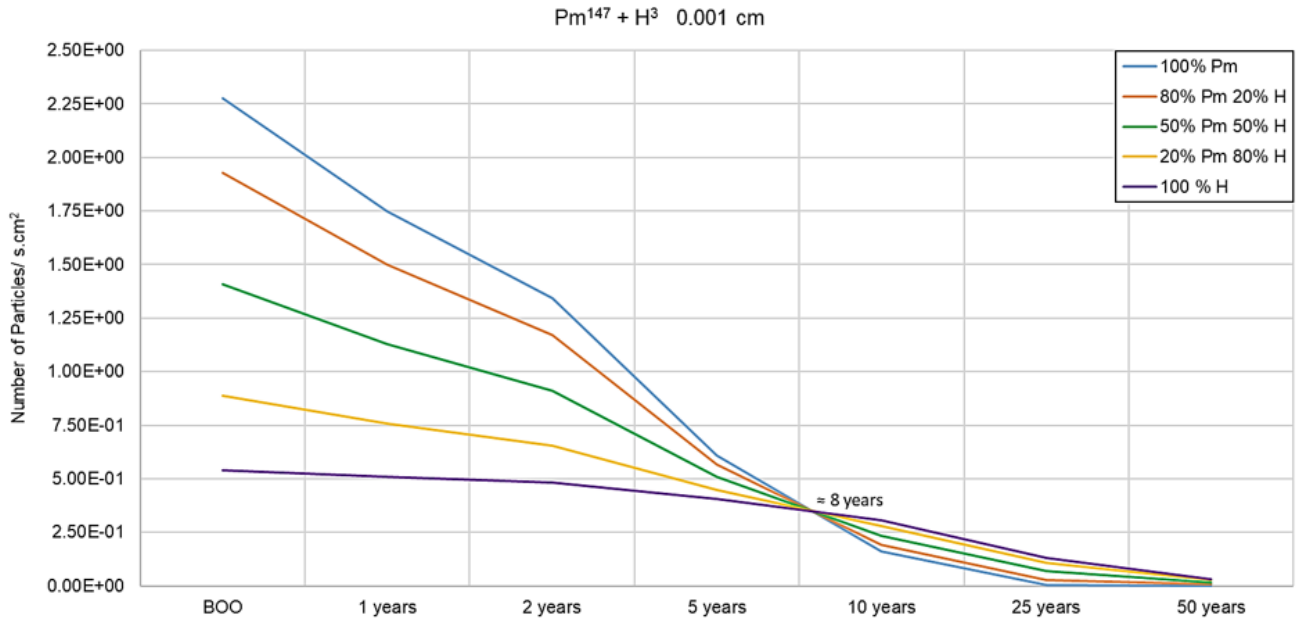


Fig. 5: Pm^{147} and H^3 Battery mix battery graph at number of particles generated in a time lapse. Source thickness was 0.001 cm.

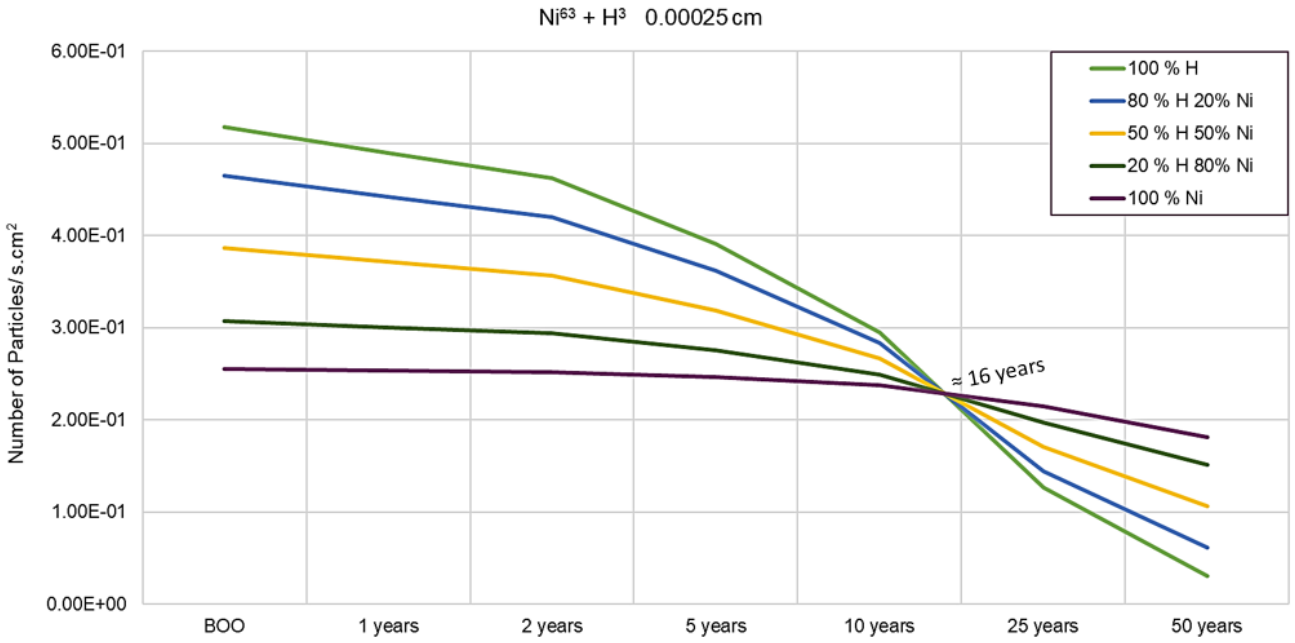


Fig. 6: Ni^{63} and H^3 Battery mix battery graph at number of particles generated in a time lapse. Source thickness was 0.00025 cm.

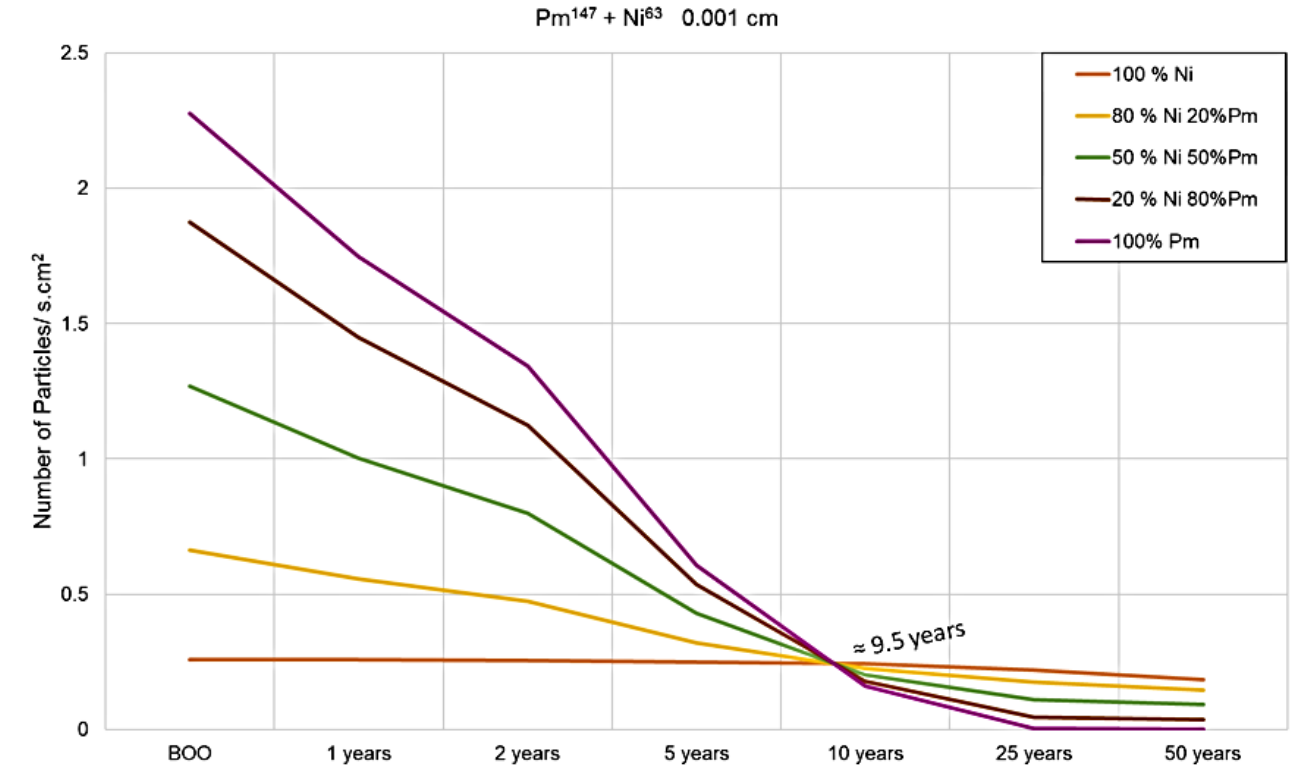


Fig. 7: Ni^{63} and Pm^{147} Battery mix battery graph at number of particles generated in a time lapse. Source thickness was 0.001 cm.

5. Discussion

a) Pm^{147} and H^3 Battery mix;

For example, at the BOO a source containing 20% Pm^{147} and 80% H^3 will emit 1.64x more particles than pure H^3 and 2.56 less particles than pure Pm^{147}. At 50 years, the 20% Pm^{147} and 80% H^3 will emit 6317x more particles than pure Pm^{147} and 1.25x less particles than pure H^3. 0.001 cm was chosen as a cutoff thickness due to being intermediate to Pm^{147} (50 μm) and H^3 (2.5 μm) cutoff thickness in regards to self-absorption. In this case, the mixed option would maybe be a good option for a device that needs more power than a H^3 pure battery for a longer time than a Pm^{147} pure battery.

b) Ni^{63} and H^3 Battery mix;

For example, at BOO a source containing 50% Ni^{63} and 50% H^3 will emit 1.34x less particles than pure H^3 and 1.51 more particles than pure Ni^{63}. At 50 years, the 50% Ni^{63} and 50% H^3 will emit 1.70x less particles than pure Ni^{63} and 3.4x more particles than pure H^3. 0.00025 cm was chosen as a cutoff thickness due to it being exactly the Ni^{63} and H^3 cutoff thickness in regards to self-absorption. In this case, the difference is not so drastic, but it might be a good option when considering the high Ni^{63} costs.

c) Ni^{63} and Pm^{147} Battery mix;

For example, at BOO a source containing 50% Pm-147 and 50% Ni-63 will emit 4.89x more particles than pure Ni-63 and 1.78 less particles than pure Pm-147. At 50 years, the 50% Pm-147 and 50% Ni-63 will emit 21951x more particles than pure Pm-147 and 2.04x less particles than pure Ni-63. 0.001 cm was chosen as a cutoff thickness due to being intermediate to Pm-147 (50 μm) and Ni-63 (2.5 μm) cutoff thickness in regards to self-absorption. In this case, the mix option also showed higher shelf life.

6. Conclusion

- Pm^{147} and H^3 battery: there is an improvement in incorporating 20% of Pm^{147} in a tritium battery;
- Ni^{63} and H^3 battery: A mix Ni^{63} and H^3, has larger power at BOO and little lost after 50 years;
- Pm-147 and Ni-63 battery: there is a tremendous improvement in incorporating 20% of Pm^{147} in a Ni^{63} battery due to a high increase in power at BOO and less power loss in 50 years.

This opens a possibility of uses. Depending on the power needs and usage time, it might be a good idea to mix radio isotopes in a nuclear battery. Also considering the price to be **Pm^{147}>>>>>>>>>> Ni^{63}>>> H^3** it might be a interesting strategy to cut costs.

REFERENCES

[1] Prelas MA, Boraas M, Aguilar FDLT, Seelig J-D, Tchouaso MT, Wisniewski D. *Nuclear Batteries and Radioisotopes*, 1st ed. ed. Switzerland: Springer International Publishing, 2016
[2] Bykov AS, Malinkovich MD, Kubasov IV, Kislyuk AM, Kiselev DA, Ksenich SV, et al. Application of Radioactive Isotopes for Beta-Voltaic Generators. *Russian Microelectronics* 2017;46:527-539
[3] Oregon State University - College of Engineering. Radioisotopes Powering Undersea Research. Accessed 15 May, 2019
[4] Summerer L, Stephenson K. Nuclear Power Sources: A Key Enabling Technology for Planetary Exploration. *Proceedings of the Institution of Mechanical Engineers, Part G: Journal of Aerospace Engineering* 2011;225:129-143
[5] Bass JC, Allen DT. Milliwatt radioisotope power supply for space applications. In:*Eighteenth International Conference on Thermoelectrics. Proceedings, ICT'99 (Cat. No.99TH8407)*, 1999; 521-524
[6] Prelas MA, Weaver CL, Watermann ML, Lukosi ED, Schott RJ, Wisniewski DA. A Review of Nuclear Batteries. *Progress in Nuclear Energy* 2014;75:117-148
[7] Daruich de Souza C, Kim JB, Kim JJ, Kim J, Ji W, Son KJ, et al. The Basics of Betavoltaic Nuclear Batteries. *International Journal of Current Advanced Research* 2021;in press
[8] Broderick K, Lusk R, Hinderer J, Griswold J, Boll R, Garland M, et al. Reactor production of promethium-147. *Applied Radiation and Isotopes* 2019;144:54-63
[9] Hinderer JH. Radioisotopic Impurities in Promethium-147 Produced at the ORNL High Flux Isotope Reactor. In.

https://trace.tennessee.edu/utk_gradthes/717: University of Tennessee, 2010
[10] International Atomic Energy Agency. Manual for reactor produced radioisotopes. In. Vienna: International Atomic Energy Agency, 2003
[11] Andreev VM, Kevetsky AG, Kaiinovsky VS, Khvostikov V, Larionov VR, Rumyantsev VD, et al. *Tritium-powered betacells based on AlxGa1-xAs*. In: IEEE, ed. *Twenty-Eighth IEEE Photovoltaic Specialists Conference Book* Piscataway, New Jersey, USA: Institute of Electrical and Electronics Engineers, 2000:1253-1256
[12] Deus S. Tritium-powered betavoltaic cells based on amorphous silicon. In:*28th IEEE Photovoltaic Specialists Conference*. Anchorage, USA, 2000; 1246-1249
[13] Royal Society of Chemistry. Aluminium. Accessed Oct. 30 2021
[14] Chikalla TD, McNeilly CE, Roberts FP. Polymorphic Modifications of Pm2O3. *Journal of the American Ceramic Society* 1972;55:428-428
[15] Royal Society of Chemistry. Nickel. Accessed Dec. 6, 2021
[16] Water Science School USGS. Water Density. Accessed Oct. 30, 2021
[17] McConn Jr RJ, Gesh CJ, Pagh RT, Rucker RA, Williams III RG. Compendium of Material Composition Data for Radiation Transport Modeling. In. 1 ed ed. Oak Ridge, Tn, USA. https://www.pnnl.gov/main/publications/external/technical_reports/PNNL-15870Rev1.pdf: U.S. Department of Homeland Security, 2011; 375