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Typical Neutron Emission Spectra for Multi-Mission Radioisotope Thermoelectric Generator Fuel

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Changing the World's Energy Future

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Idaho National Laboratory

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1.0 Background

The Dragonfly [1] rotorcraft currently being designed by the Johns Hopkins Applied Physics Laboratory (APL) is a mission destined to explore, via autonomous flight, the Saturnian moon of Titan and currently scheduled to launch in 2027. This largest moon of Saturn contains a thick, dense atmosphere, that when coupled with the remote distance to the Sun, requires the use of a radioisotope power system (RPS). The multi-mission radioisotope thermoelectric generator (MMRTG) fueled at Idaho National Laboratory is currently the only flight-certified RPS still in production within the Department of Energy complex, thus, an MMRTG was chosen for the Dragonfly mission.

2.0 Purpose

One of the many instruments being incorporated into Dragonfly, the Dragonfly Gamma-Ray and Neutron Spectrometer (DRaGNS) consists of a high-purity germanium detector that is sensitive to damage caused by high energy neutrons. The MMRTG fuel is plutonium-238 (^{238}Pu) in an oxide form (PuO_2) that emits neutrons through many different modes, however the primary modes are via spontaneous fission (SF) and alpha-neutron (α, n) reactions with the light elements present in the fuel [2]. Per Taherzadeh [2], the neutrons emitted via spontaneous fission can be represented by the energy dependent equation:

$$N(E) = 2.04 \times 10^3 (E)^{0.5} e^{\left(\frac{-E}{1.34}\right)}$$

in n/s/MeV/g^{238}Pu, where E is the neutron energy in MeV. However, the number of low-Z impurities in the fuel as well as the amount of ^{18}O present in the oxides bonded to the plutonium can significantly alter the portion of the neutron spectrum caused by (α, n) reactions, and because the composition of the fuel has undergone significant variation since the neutron spectrum measurement was taken at Mound in 1985 [3], a more modern representation of the spectrum was needed. Though a more recent estimate of the neutron spectrum was published by I. Jun et al. in 2013 [4] via direct measurements of the neutron background on the surface of Mars, this spectrum did not include neutrons with energies greater than 100 keV and used an inferred fuel composition. Neutron irradiation damage is proportional to neutron energy, so having a published spectrum representative of the most recent fuel composition, as well as one that incorporates neutrons of all relevant emitted energies, is imperative for the Dragonfly mission. It is also of note that future RPS-powered missions will need such data if they utilize neutron-sensitive instruments.

3.0 Fuel Neutron Spectrum Data

In order to produce this data, the approach was that of generating a simulated neutron spectrum emitted by the fuel, based on its known composition. This differs from the measured data [3] [4], which includes subsequent neutron scattering and absorption in materials within the generator as well as induced fissions within the fuel. To generate such a simulated neutron spectrum, the radioisotope power system dose estimation tool (RPS-DET) software [5][6] was used. RPS-DET is a graphical user interface for the SCALE software suite that can, amongst other capabilities, conveniently execute ORIGEN to generate time-dependent neutron and gamma source spectra for a given RPS fuel composition [7].

An initial plutonium isotopic composition was assumed to be the default composition incorporated into RPS-DET and is shown in Table 3.1. Because there has been non-trivial variation in low-Z impurities, ^{18}O, and corresponding neutron emission rates in the batches made during the campaigns for the Curiosity and Perseverance rovers, three initial impurity compositions were assumed and defined in Table 3.2 to provide readers with appropriate low, high, and mid-range neutron intensities and spectra to be used for modeling, design, and research purposes concerning contemporary spaceflight PuO_2.

Table 3.1. Plutonium Isotopic Fuel Composition

Component	wt% in PuO_2
O and impurities	13.00%
^{238}Pu	74.80%
^{239}Pu	9.19%
^{240}Pu	2.66%
^{241}Pu	0.06%
^{242}Pu	0.01%

Table 3.2. Description of Assumed Impurity Compositions

Impurity Composition	Description
1	Average of the low-Z impurities and neutron emission rates of the most recent MMRTG fuel clads (as of August 2022)
2	Default amount of low-Z impurities and ^{18}O in RPS-DET
3	Maximum of low-Z fuel impurities allowed per fuel specification and default amount of ^{18}O in RPS-DET

The plutonium assay between all three compositions is the same, but the amount of ^{18}O in composition 1 is representative of the average neutron emission rate observed in the thirty-six most recent fuel clads (as of August 2022). Composition 2 corresponds to the default impurities and ^{18}O amounts assumed in RPS-DET, while composition 3 maintains the same ^{18}O amount as Composition 2 but includes all other impurities at their maximum allowed limits. The energy dependent neutron data for each composition at beginning of life is plotted in Figure 3.1. The reader should keep in mind, these spectra are representative of fresh fuel clads at beginning of life (i.e., less than one year in age) and only the pure emission spectrum (both spontaneous fission and α,n).

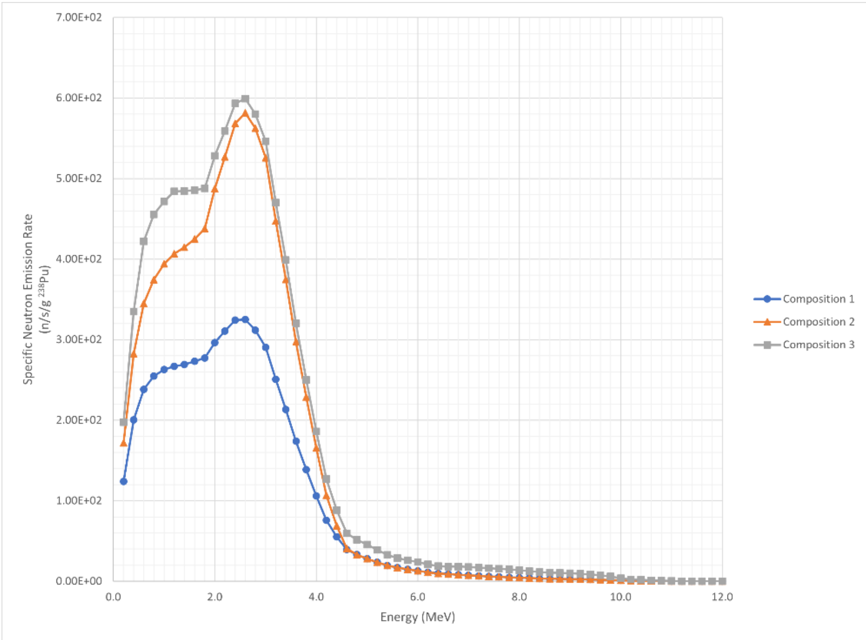

Figure 3.1. Comparison of Composition Dependent MMRTG Fuel Neutron Spectra at Beginning of Life

Though the total neutron emission rate for each fuel composition does scale with the radioactive decay of ^{238}Pu, time dependent data is plotted for convenience in Figure 3.2. The Data Appendix contains the tabulated data for Figure 3.1 and Figure 3.2 in Table 8.1 and Table 8.2, respectively.

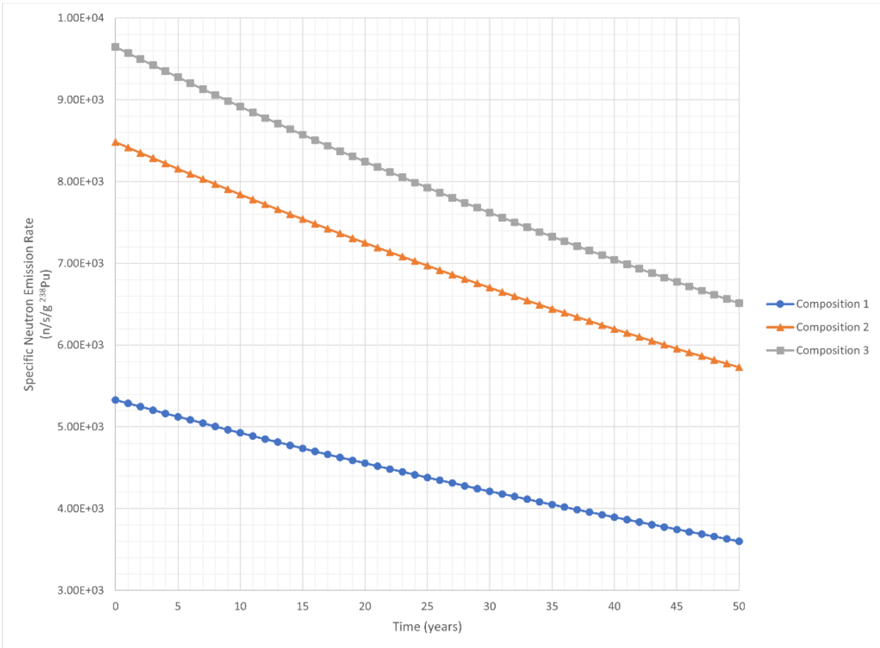

Figure 3.2. Time Dependent Behavior of MMRTG Fuel Total Neutron Emission Rate for each Fuel Composition

4.0 Data Analysis

Because the low-Z impurities in composition 1 match the average of the most recent fuel produced, this neutron spectrum can be considered representative of nominal contemporary PuO_2 fuel. Composition 2 can be considered an intermediate fuel and composition 3 an unlikely worst-case scenario fuel. The reader should keep in mind, these spectra are representative of fresh fuel clads at beginning of life (i.e., less than one year in age) and only the pure emission spectrum (both spontaneous fission and α,n). These spectra do not account for any subsequent neutron scattering or induced fissions in materials like fuel, cladding, insulation, or other MMRTG components. This is important when considering the spectrum that was measured by Mound was that of a general purpose heat source radioisotope thermoelectric generator (GPHS-RTG) [8] and the partial spectrum measured by the Curiosity Rover instrument was that of a MMRTG, not the fuel alone. The compositional differences between the two designs of generators, particularly the unicouple materials and the amount of fuel, could contribute to non-trivial differences in the effective neutron field and is a consideration for future work.

5.0 Conclusions

The measured data from Mound was a good representation of the neutron field surrounding a GPHS-RTG, however the differences in the MMRTG design and the non-trivial variation in low-Z impurities of the fuel prompted the publication of a revised spectrum indicative of recent fuel compositions. The simulated neutron spectrum for each fuel composition plotted in Figure 3.1 represents a nominal contemporary fuel with composition 1 and then increased conservatism with composition 2, culminating with an unlikely worst-case scenario of composition 3. Considering the spectrum of composition 1 is derived from the average of just over thirty-two fuel clads, it is a good representation of the neutron spectrum for the fuel in a full MMRTG. If a spectrum is needed for a conservative shielding design, the composition 3 spectrum could be considered appropriate.

6.0 Acknowledgements

The author would like to thank Nathan Gilliam of Idaho National Laboratory (INL) for acquiring the impurity data for the most recent fuel, Michael Smith of Oak Ridge National Laboratory for contributions to content editing and guidance on RPS-DET, and Eric Clarke of INL as well for his guidance on content and scope of this document. The author would also like to thank Brad Kirkwood for his valued contributions on this and previous collaborations regarding MMRTG fuel compositions and radiation field analyses.

7.0 References

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8.0 Data Appendix

Table 8.1. MMRTG Fuel Neutron Source Spectrum Data at Beginning of Life

Energy (MeV)	Impurity Composition		
	1	2	3
	Specific Neutron Emission Rate (n/s/g ^{238}Pu)		
0.2	1.24E+02	1.72E+02	1.98E+02
0.4	2.00E+02	2.82E+02	3.35E+02
0.6	2.38E+02	3.45E+02	4.22E+02
0.8	2.55E+02	3.74E+02	4.56E+02
1.0	2.63E+02	3.94E+02	4.72E+02
1.2	2.67E+02	4.07E+02	4.84E+02
1.4	2.69E+02	4.15E+02	4.84E+02
1.6	2.73E+02	4.25E+02	4.86E+02
1.8	2.77E+02	4.38E+02	4.88E+02
2.0	2.96E+02	4.87E+02	5.28E+02
2.2	3.11E+02	5.27E+02	5.59E+02
2.4	3.24E+02	5.68E+02	5.93E+02
2.6	3.25E+02	5.82E+02	5.99E+02
2.8	3.12E+02	5.62E+02	5.80E+02

Energy (MeV)	Impurity Composition		
	1	2	3
	Specific Neutron Emission Rate (n/s/g ^{238}Pu)		
3.0	2.91E+02	5.25E+02	5.46E+02
3.2	2.51E+02	4.47E+02	4.70E+02
3.4	2.13E+02	3.75E+02	3.99E+02
3.6	1.74E+02	2.97E+02	3.20E+02
3.8	1.39E+02	2.29E+02	2.50E+02
4.0	1.06E+02	1.66E+02	1.86E+02
4.2	7.58E+01	1.07E+02	1.27E+02
4.4	5.53E+01	6.88E+01	8.84E+01
4.6	3.93E+01	4.04E+01	5.96E+01
4.8	3.31E+01	3.30E+01	5.16E+01
5.0	2.82E+01	2.81E+01	4.57E+01
5.2	2.36E+01	2.35E+01	3.90E+01
5.4	1.97E+01	1.97E+01	3.30E+01
5.6	1.70E+01	1.70E+01	2.86E+01

Energy (MeV)	Impurity Composition		
	1	2	3
	Specific Neutron Emission Rate (n/s/g ^{238}Pu)		
5.8	1.49E+01	1.49E+01	2.62E+01
6.0	1.31E+01	1.31E+01	2.40E+01
6.2	1.13E+01	1.13E+01	2.09E+01
6.4	9.84E+00	9.82E+00	1.87E+01
6.6	8.88E+00	8.85E+00	1.82E+01
6.8	8.15E+00	8.12E+00	1.83E+01
7.0	7.39E+00	7.36E+00	1.77E+01
7.2	6.68E+00	6.65E+00	1.70E+01
7.4	6.06E+00	6.03E+00	1.63E+01
7.6	5.51E+00	5.49E+00	1.56E+01
7.8	4.98E+00	4.96E+00	1.48E+01
8.0	4.47E+00	4.45E+00	1.38E+01
8.2	3.99E+00	3.97E+00	1.26E+01
8.4	3.57E+00	3.55E+00	1.16E+01
8.6	3.23E+00	3.22E+00	1.09E+01
8.8	2.96E+00	2.94E+00	1.03E+01
9.0	2.74E+00	2.72E+00	9.87E+00
9.2	2.53E+00	2.52E+00	9.44E+00
9.4	2.29E+00	2.27E+00	8.70E+00
9.6	1.96E+00	1.94E+00	7.48E+00
9.8	1.56E+00	1.55E+00	5.86E+00
10.0	1.10E+00	1.10E+00	3.90E+00
10.2	7.68E-01	7.63E-01	2.48E+00
10.4	5.98E-01	5.95E-01	1.84E+00
10.6	4.56E-01	4.54E-01	1.32E+00
10.8	3.26E-01	3.25E-01	8.27E-01
11.0	2.13E-01	2.13E-01	3.94E-01
11.2	1.42E-01	1.42E-01	1.46E-01
11.4	1.18E-01	1.18E-01	1.18E-01
11.6	9.89E-02	9.89E-02	9.89E-02
11.8	8.29E-02	8.29E-02	8.29E-02
12.0	6.95E-02	6.95E-02	6.95E-02
12.2	5.82E-02	5.82E-02	5.82E-02
12.4	4.88E-02	4.88E-02	4.88E-02
12.6	4.08E-02	4.08E-02	4.08E-02
12.8	3.41E-02	3.41E-02	3.41E-02
13.0	2.85E-02	2.85E-02	2.85E-02

Energy (MeV)	Impurity Composition		
	1	2	3
	Specific Neutron Emission Rate (n/s/g ^{238}Pu)		
13.2	2.39E-02	2.39E-02	2.39E-02
13.4	1.99E-02	1.99E-02	1.99E-02
13.6	1.66E-02	1.66E-02	1.66E-02
13.8	1.39E-02	1.39E-02	1.39E-02
14.0	1.16E-02	1.16E-02	1.16E-02
14.2	9.67E-03	9.67E-03	9.67E-03
14.4	8.06E-03	8.06E-03	8.06E-03
14.6	6.72E-03	6.72E-03	6.72E-03
14.8	5.60E-03	5.60E-03	5.60E-03
15.0	4.66E-03	4.66E-03	4.66E-03
15.2	3.88E-03	3.88E-03	3.88E-03
15.4	3.23E-03	3.23E-03	3.23E-03
15.6	2.69E-03	2.69E-03	2.69E-03
15.8	2.24E-03	2.24E-03	2.24E-03
16.0	1.86E-03	1.86E-03	1.86E-03
16.2	1.55E-03	1.55E-03	1.55E-03
16.4	1.28E-03	1.28E-03	1.28E-03
16.6	1.07E-03	1.07E-03	1.07E-03
16.8	8.86E-04	8.86E-04	8.86E-04
17.0	7.35E-04	7.35E-04	7.35E-04
17.2	6.10E-04	6.10E-04	6.10E-04
17.4	5.06E-04	5.06E-04	5.06E-04
17.6	4.20E-04	4.20E-04	4.20E-04
17.8	3.48E-04	3.48E-04	3.48E-04
18.0	2.89E-04	2.89E-04	2.89E-04
18.2	2.39E-04	2.39E-04	2.39E-04
18.4	1.98E-04	1.98E-04	1.98E-04
18.6	1.64E-04	1.64E-04	1.64E-04
18.8	1.36E-04	1.36E-04	1.36E-04
19.0	1.13E-04	1.13E-04	1.13E-04
19.2	9.32E-05	9.32E-05	9.32E-05
19.4	7.71E-05	7.71E-05	7.71E-05
19.6	6.38E-05	6.38E-05	6.38E-05
19.8	5.27E-05	5.27E-05	5.27E-05
20.0	4.36E-05	4.36E-05	4.36E-05
Total	**5.33E+03**	**8.48E+03**	**9.65E+03**

Table 8.2. Time Dependent MMRTG Fuel Neutron Emission Rate Data

Time (years)	Impurity Composition		
	1	2	3
	Specific Neutron Emission Rate (n/s/g ^{238}Pu)		
0	5.33E+03	8.48E+03	9.65E+03
1	5.29E+03	8.42E+03	9.57E+03
2	5.25E+03	8.35E+03	9.50E+03
3	5.21E+03	8.29E+03	9.42E+03
4	5.17E+03	8.22E+03	9.35E+03
5	5.12E+03	8.16E+03	9.28E+03
6	5.08E+03	8.09E+03	9.20E+03
7	5.05E+03	8.03E+03	9.13E+03
8	5.01E+03	7.97E+03	9.06E+03
9	4.97E+03	7.90E+03	8.99E+03
10	4.93E+03	7.84E+03	8.92E+03
11	4.89E+03	7.78E+03	8.85E+03
12	4.85E+03	7.72E+03	8.78E+03
13	4.81E+03	7.66E+03	8.71E+03
14	4.78E+03	7.60E+03	8.64E+03
15	4.74E+03	7.54E+03	8.57E+03
16	4.70E+03	7.48E+03	8.51E+03
17	4.66E+03	7.42E+03	8.44E+03
18	4.63E+03	7.37E+03	8.37E+03
19	4.59E+03	7.31E+03	8.31E+03
20	4.56E+03	7.25E+03	8.24E+03
21	4.52E+03	7.19E+03	8.18E+03
22	4.49E+03	7.14E+03	8.12E+03
23	4.45E+03	7.08E+03	8.05E+03
24	4.42E+03	7.03E+03	7.99E+03
25	4.38E+03	6.97E+03	7.93E+03
26	4.35E+03	6.92E+03	7.86E+03
27	4.31E+03	6.86E+03	7.80E+03
28	4.28E+03	6.81E+03	7.74E+03
29	4.25E+03	6.76E+03	7.68E+03
30	4.21E+03	6.70E+03	7.62E+03
31	4.18E+03	6.65E+03	7.56E+03
32	4.15E+03	6.60E+03	7.50E+03
33	4.12E+03	6.55E+03	7.44E+03
34	4.08E+03	6.50E+03	7.38E+03
35	4.05E+03	6.44E+03	7.33E+03
36	4.02E+03	6.39E+03	7.27E+03
37	3.99E+03	6.34E+03	7.21E+03
38	3.96E+03	6.29E+03	7.16E+03
39	3.93E+03	6.25E+03	7.10E+03
40	3.90E+03	6.20E+03	7.04E+03
41	3.87E+03	6.15E+03	6.99E+03
42	3.84E+03	6.10E+03	6.94E+03
43	3.81E+03	6.05E+03	6.88E+03
44	3.78E+03	6.00E+03	6.83E+03
45	3.75E+03	5.96E+03	6.77E+03
46	3.72E+03	5.91E+03	6.72E+03
47	3.69E+03	5.87E+03	6.67E+03
48	3.66E+03	5.82E+03	6.62E+03
49	3.63E+03	5.77E+03	6.56E+03
50	3.60E+03	5.73E+03	6.51E+03