ALTERNATIVE FUEL SOURCES FOR RADIOISOTOPE THERMOELECTRIC GENERATORS

An Undergraduate Research Scholars Thesis

by

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ABSTRACT

Alternative Fuel Sources for Radioisotope Thermoelectric Generators. (May 2015)

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With Pu-238 in short supply, it is necessary to investigate other potential fuel sources that may be suitable for powering radioisotope thermoelectric generators (RTGs). This report has investigated several candidate isotopes based on their power output, decay products, half-lives, shielding requirements, availability and ability to be isolated. Through manipulation of the SCALE modeling program, the feasibility of using transuranic radioisotopes found in spent fuel was thoroughly examined. In addition to examining the use of single isotopes, the possibility of selecting an isotope that will decay into another feasible isotope will be explored. This report attempts to identify isotopes that are suitable alternatives to Pu-238 that both fulfill the power requirements of deep space missions and stay within the economic constraints associated with such projects.

DEDICATION

To Mom and Joe, who have provided me with more than any son deserves. I've got a long ways to go, but I know that you'll be there every step of the way.

-Evan

ACKNOWLEDGEMENTS

We would like to thank our advisor, Dr. Pavel Tsvetkov, for his continual guidance and help throughout this project. Also, we would like to thank to our colleagues who have recently joined the project; we look forward to working with you.

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Thanks to ORNL for providing the software used to perform analysis throughout the project.

NOMENCLATURE

RTG	Radioisotope Thermoelectric Generator
ORNL	Oak Ridge National Laboratory
OECD	Organization for Economic Co-operation and Development
NEA	Nuclear Energy Agency
JANIS	Java-based Nuclear Data Information System
SCALE	Standardized Computer Analyses for Licensing Evaluation
CANDU	Canada Deuterium Uranium
GE	General Electric

CHAPTER I

BACKGROUND

Importance of RTGs

One of the most reliable sources of power employed by deep-space probes today is the radioisotope thermoelectric generator (RTG). By the conversion of thermal energy produced from radioactive decay into electricity by means of thermoelectric generation, RTG's are able to operate independent of moving parts and continuous maintenance. Most RTGs employed by NASA for deep-space missions are powered by Pu-238, shown in Figure 1. However, the transition from Pu-238 to an alternative radioisotope for powering RTGs is becoming increasingly necessary as global stockpiles of Pu-238 decline. Current stockpiles were derived primarily as a byproduct in the creation of weapons grade Pu-239, however, production was suspended in the United States after 1986. Even with the recent restart of production (roughly 1.5 kg/year) at Oak Ridge National Laboratory (ORNL) [1], it is apparent that there is more demand for RTG fuel than is currently available. This means that in order to continue utilizing explorative technology such as the Curiosity rover or the Voyager spacecraft, a viable alternative must implemented.

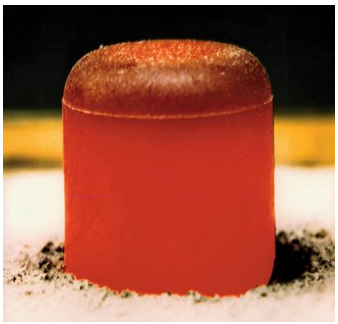

Figure 1: Pu-238 Oxide Fuel Pellet [2]

Working Mechanism

Thermoelectric generation takes advantage of the Seebeck effect, in which a voltage differential is generated through the junction between two wires of differing metals which experience a temperature gradient between their opposing ends. With the use of both p-type and n-type semiconductors (having crystal lattices allowing the flow of positive and negative charges, respectively), higher temperatures and efficiencies can be reached, allowing for useful amounts of electric power to be generated. An illustration of this effect is shown in Figure 2.

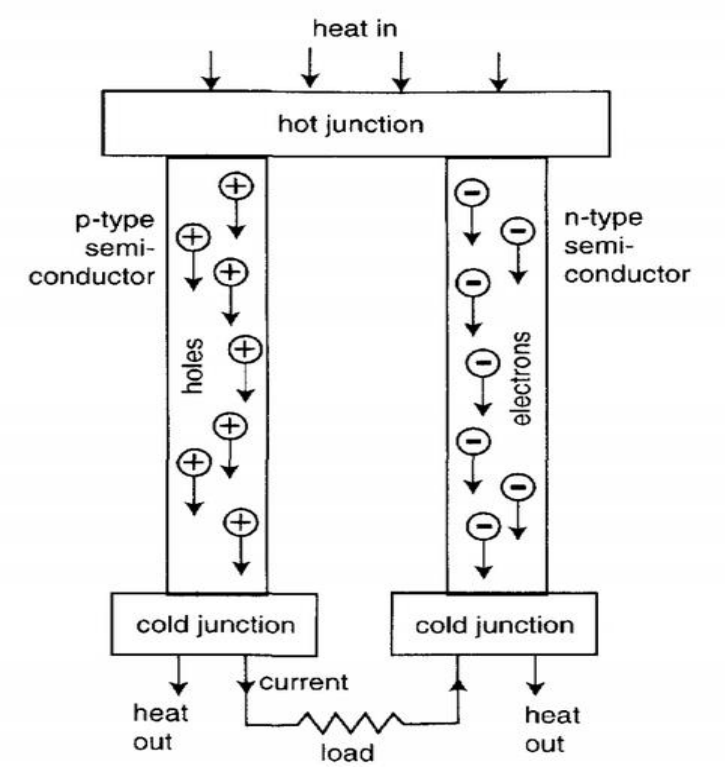

Figure 2: A thermoelectric converter cell [3]

The typical efficiency of an RTG ranges from 1-10%, producing 0.3-1.2 V per cell, although there is currently work being done to improve these efficiencies. In order to attain higher voltages and greater power, several converter cells are usually connected in series [3]. A typical configuration is shown in Figure 3.

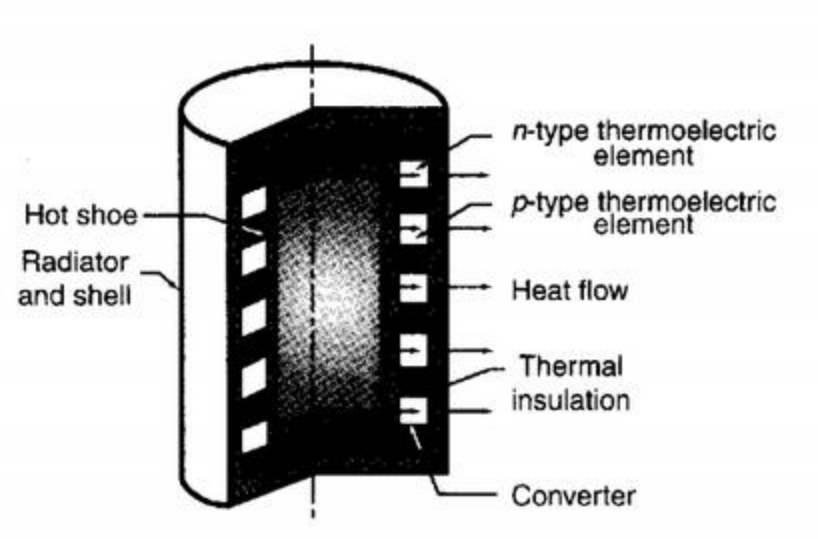

Figure 3: A thermoelectric isotopic power generator [3]

Previous Uses of RTGs

Since 1961, the U.S. has launched 26 space missions that have carried over 40 RTGs to provide part or all of the electrical power needs of the instrumentation on board. These RTGs had electrical power capacities ranging from a few W(e) up to 285 W(e) [3]. Many of the early RTGs used in these missions are listed in Table 1.

Table 1: Early (1960-1975) U.S. SNAP radioisotope power generators [3]

SNAP No.	Function	Fuel	Power (We)	Dia. × Ht. (cm)	Mass (kg)	Design Life
3	demonstration	^{210}Po	2.5	12×14	1.82	90 d
3A	satellite power	^{238}Pu	2.7	12×14	2.10	5 y
-	weather station	^{90}Sr	5	46×51	764	2 y min
7A	navigation buoy	^{90}Sr	10	51×53	850	2 y min
7B	navigation light	^{90}Sr	60	56×88	2100	2 y min
7C	weather station	^{90}Sr	10	51×53	850	2 y min
7D	floating weather station	^{90}Sr	60	56×88	2100	2 y min
7E	ocean bottom beacon	^{90}Sr	7.5	51×53	273	2 y min
7F	offshore oil rig	^{90}Sr	60	56×88	2100	2 y min
9A	satellite power	^{238}Pu	25	51×24	12	5 y
11	moon probe	^{242}Cm	23	51×30	14	90 d
13	demonstration	^{242}Cm	12	6.4×10	1.8	90 d
15	military	^{238}Pu	0.001	7.6×7.6	0.5	5 y
17	communication satellite	^{90}Sr	25	61×36	14	5 y
19	Nimbus weather satellites	^{238}Pu	30	56×25	14	5 y
19	Viking/Pioneer missions	^{238}Pu	45			5 y
21	deep sea application	^{90}Sr	10	41×61	230	5 y
23	terrestrial uses	^{90}Sr	60	64×64	410	5 y
27	Apollo lunar modules	^{238}Pu	60	46×46	14	5 y
29	various missions	^{210}Po	500		230	90 d

CHAPTER II

INTRODUCTION

In order to satisfy the need for an alternative fuel source, candidate radioisotopes capable of powering an RTG will need to be identified; a safety analysis of the use of this isotope in RTGs will be necessary as well. In the event of re-entry, launch failure, or various other extreme test conditions, the chosen radioisotope must not pose any serious hazards. Containment of the fuel during any of these events is crucial, and so possible containment configurations and materials would ideally be examined as well. Possible fuel assembly designs have been hypothesized by Ambrosi at the Nuclear and Emerging Technologies for Space Conference (2012) [4]. Preliminary research has shown that Am-241, Cm-242, Po-210 and a handful of other isotopes are prime candidates for fulfilling the aforementioned requirements established for a suitable radioisotope, and their properties will be further investigated.

The purpose of this paper is to compare the various alternative fuel candidates, designate the most efficient of these to be used in future RTGs, and deliver a thorough reasoning as to why it was selected for further considerations. Some of the primary characteristics that must be met include: a desirable half-life in order to sustain power for long term deep-space missions, a sufficient power density to meet application needs, low shielding requirements to protect equipment from radiation damage, availability of the radioisotope in spent nuclear fuel or its "manufacturability" via irradiation of parent nuclides in reactors or accelerator environments, and relative ease of isolation from other elements and isotopes. All of these characteristics will be taken into account when narrowing down the selection of isotopes to be considered as alternative fuel candidates. Through the use of the SCALE code system, developed by ORNL,

the buildup of various radioisotopes over time, as well as the power that each individual isotope produces, can be measured and interpreted for the selection process. In addition, resources such as OECD NEA JANIS can be used to analyze the properties of various candidate isotopes in order to ensure that reliable data is being used.

CHAPTER III

METHODS

Modeling and Simulations

The bulk of simulation throughout the project was done with SCALE 6.1, a nuclear safety analysis and design modeling program provided by ORNL. Specifically, ORIGEN-ARP, a graphical interface system within SCALE used to perform nuclear irradiation and decay calculations, was used to identify candidate isotopes that might be found in significant quantities within spent nuclear fuel; assuring that a suitable output would be maintained for each isotope. After specifying several variables for the fuel input, a graphical display was generated for both the mass and power outputs of various isotopes over time, running irradiation and decay cases, respectively. The simulations were run multiple times, and outputs were averaged in order to ensure a good estimate was made and that the data was not fluctuating excessively. Each isotope and their respective power outputs were recorded in Table I. The mass outputs were recorded as a percentage of the total mass of the spent fuel to allow for an easier comparison. Isotopes of favorable abundance, with respect to the abundance of other candidate isotopes, were more closely examined. In order to broaden the scope of spent fuel compositions to be examined, simulations were run using multiple reactor designs. Reactor designs included the CANDU pressurized heavy water reactor, Magnox gas cooled reactor, Westinghouse AP1000 pressurized water reactor, and GE boiling water reactor. Uranium enrichment was simulated at 3% (U -235) for all reactors, excluding the CANDU reactor, which operated at 0.711%, the abundancy of U-235 in naturally occurring uranium.

Calculating Power Densities

One of the most essential characteristics studied in the candidate isotopes was the power density they exhibited. Using ORIGEN-ARP to simulate the radioactive decay of each isotope following reactor shutdown, a direct measure of power density was derived. Power outputs from the decay simulations of each candidate isotope were recorded in watts (i.e. how much thermal power each particular isotope emitted), and then divided by their previously recorded mass in kilograms at corresponding time intervals. The result yielded a power density in units W/kg which was averaged for multiple simulations for each candidate isotope. In order to confirm the simulated results, the theoretical derivation of radioisotopic power density, also referred to as 'specific power', was applied [5].

$$P' = 1.6 \times 10^{-13} \frac{EA_v \ln 2}{MT_{1/2}} \quad (1)$$

Where: P' is power density in watts(th)/kg, E is energy release per decay event in MeV, T1/2 is the isotope half-life in seconds, M is the atomic weight in g/mol, Av is Avogadro's number = 6.022x1023 nuclei/mol, and 1.6x10-13 is a conversion factor from MeV to J.

Of course, a maximum power density is desired in selecting alternative fuel sources to RTG's. Therefore, isotopes yielding particularly low power densities were narrowed out of consideration.

Tabulated Data

In order to further investigate the potential of remaining isotopes, the JANIS database was utilized. This database served to tabulate the half-life and decay path of each nuclide, two important aspects for evaluating their use in RTG fuel. A preliminary safety analysis was also

conducted for each isotope by assessing the type of radiation prevalent in their decay, as well as in their daughter products'. This data was recorded for each nuclide and compared. High energy decay would result in excess shielding requirements; any isotopes exhibiting this type of decay were deemed less desirable.

CHAPTER IV

DISCUSSION

Generated Data

One of the benefits of using the modeling software in ORIGEN-ARP is that it allows for a variety of scenarios to be explored by varying a series of input parameters, such as the type of reactor, fuel mass and composition, period of observation, and power per respective cycle. Because the focus of this project was primarily to explore the viability of extracting candidate isotopes from spent fuel, multiple types of reactor designs and the compositions of their spent fuels were thoroughly examined. An alternative procedure for the production of fuel would be through the manufacturability of certain radionuclides through irradiation, in which case the data obtained would still be relevant apart from the measurements of mass output.

The differences in isotopic concentration among specific isotopes was shown to be significant among different reactor designs, sometimes differing several orders of magnitude. For example, the differences in concentrations of Po-210 and Cf-252 between different reactor designs can be seen below:

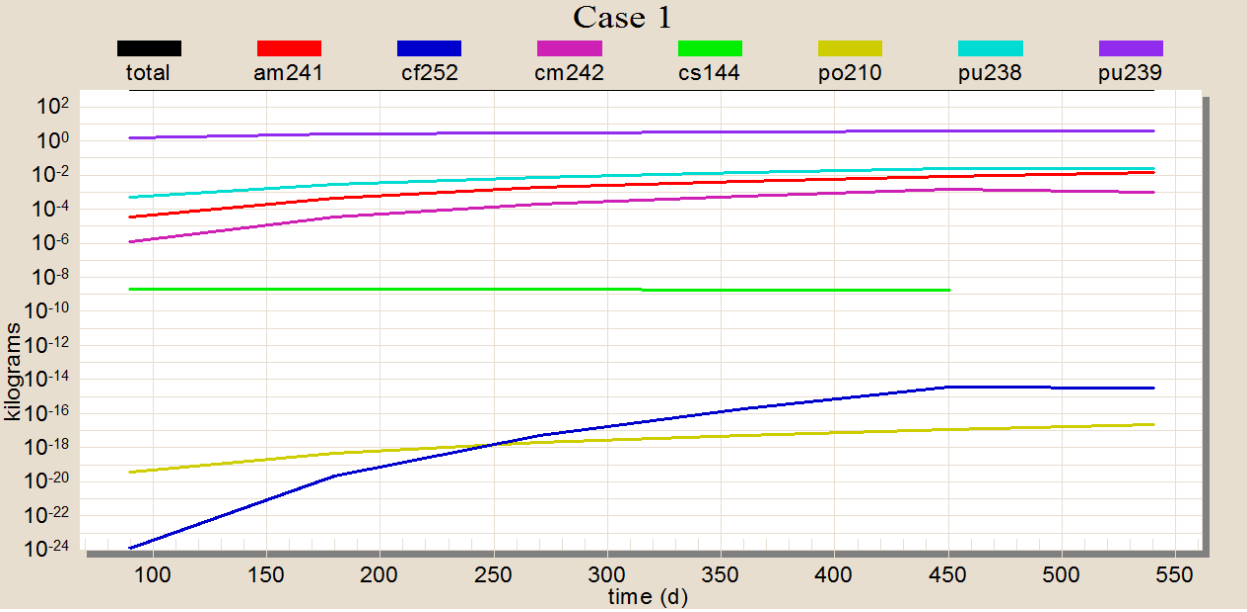

Figure 4a: Mass outputs of GE reactor

- Case Type: Irradiation
- Reactor Design: GE 10x10x8
- Observation Period:18 months (90 day cycles)
- Enrichment: 3% enriched UO_2
- Mass of Fuel: 1 Mg
- Power: 38, 40, 40, 35, 37, 0 per respective cycle

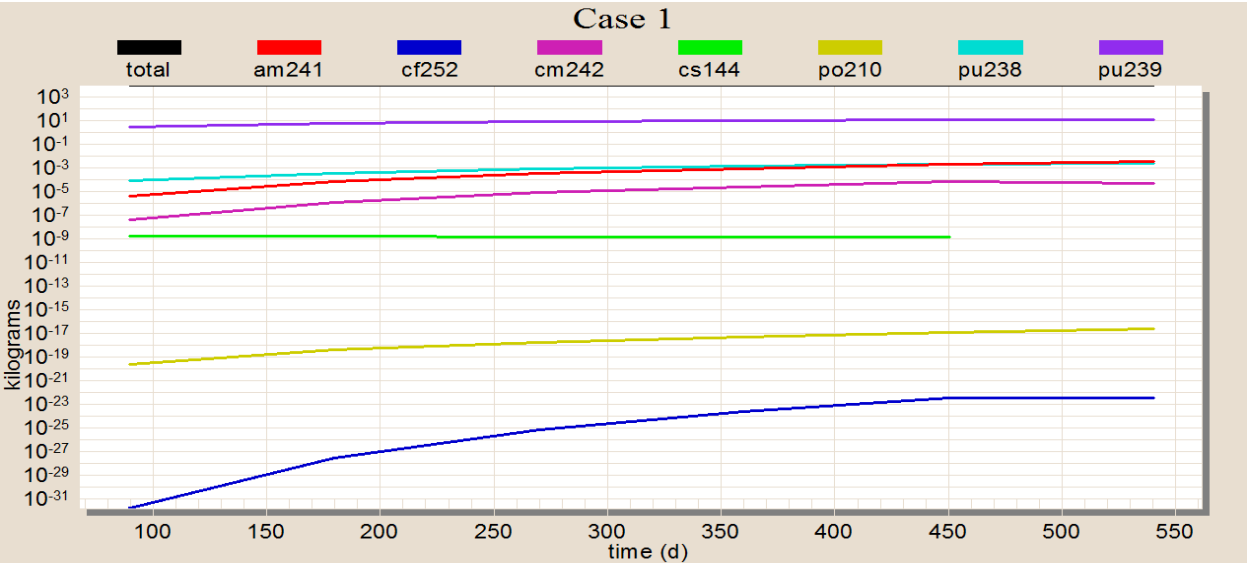

Figure 4b: Mass outputs of CANDU reactor

- Case Type: Irradiation
- Reactor Design: CANDU 28
- Observation Period: 18 months (90 day cycles)
- Enrichment: 0.711% enriched UO_2
- Mass of Fuel: 1 Mg
- Power: 38, 40, 40, 35, 37, 0 per respective cycle

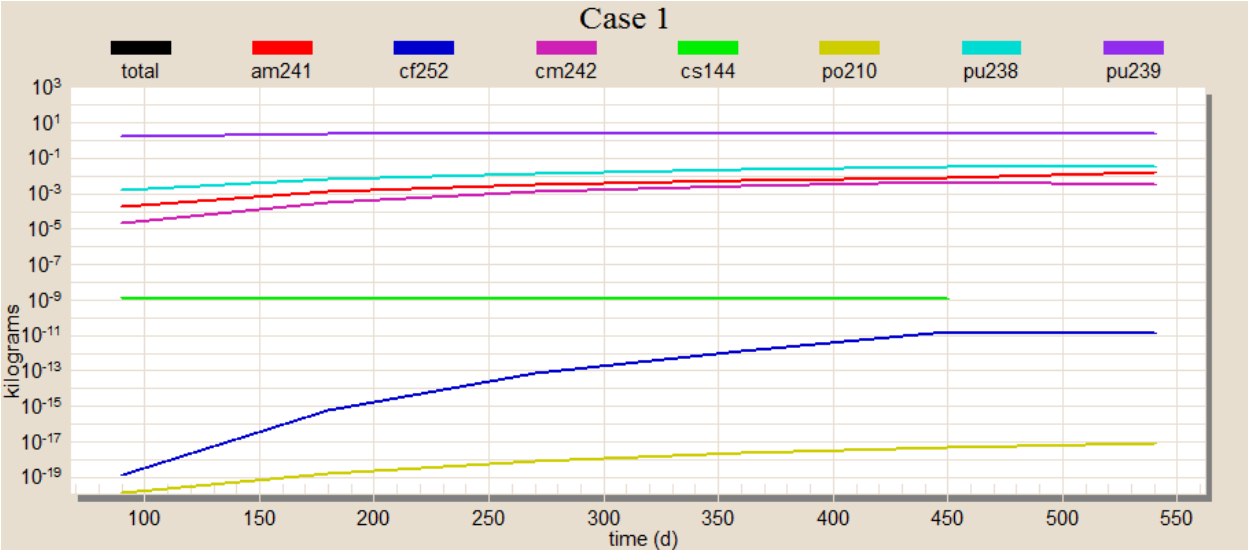

Figure 4c: Mass outputs of MAGNOX reactor

- Case Type: Irradiation
- Reactor Design: Magnox
- Observation Period: 18 months (90 day cycles)
- Enrichment: 3% enriched UO_2
- Mass of Fuel: 1 Mg
- Power: 38, 40, 40, 35, 37, 0 per respective cycle

Other useful figures generated from ORIGEN-ARP included the total power output graphs, run as a decay case. Such figures gave more context to the power potential that each isotope can provide, should it be extracted from spent fuel for use in an RTG.

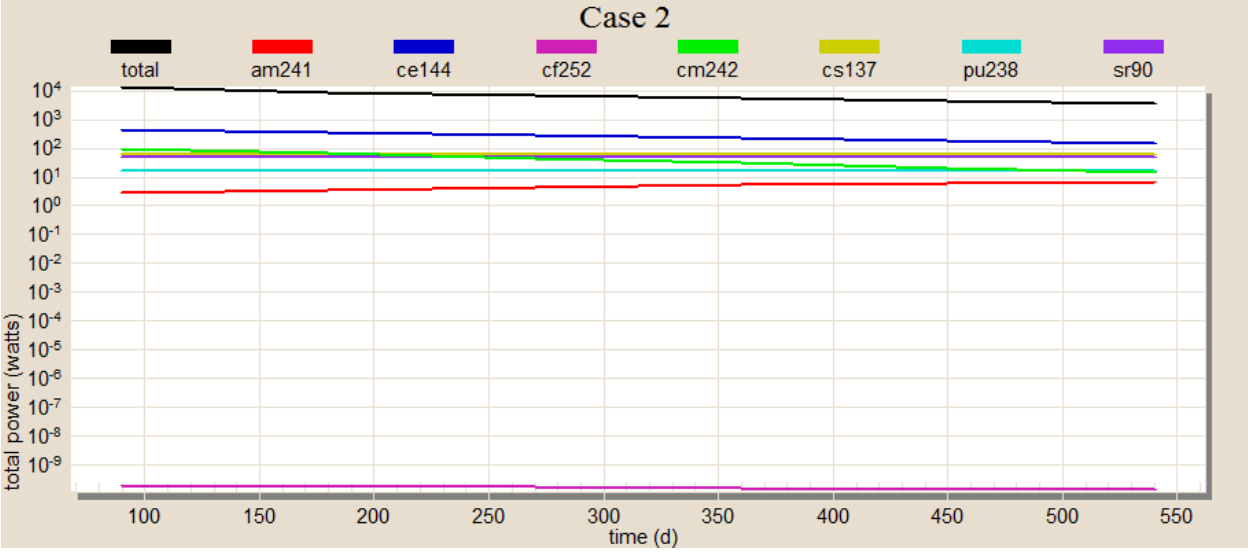

Figure 5: Power outputs of a Ce 14x14 reactor

- Case Type: Decay
- Reactor Design: Ce 14x14
- Observation Period: 18 months (90 day cycles)
- Enrichment: 3% enriched UO2
- Mass of Fuel: 1 Mg
- Power = 38, 40, 40, 35, 37, 0 per respective cycle

A review of the data has shown that although several isotopes exhibit favorable properties for use in RTG fuel, no one isotope satisfies all of the fuel requirements alone. Most notably, it has been found that where one isotope excels in power density, it lacks in half life, and vice versa. Selection of a viable fuel alternative is therefore largely determined by the specifications required by each particular mission. However, it is believed that with the advancement of next generation RTGs and their efficiency, certain isotopes may prove to be just as effective in powering deep space missions as Pu-238. The most conspicuous of these nuclides is Am-241, which boasts a 430 year half-life and 100 watts/kilogram power density, roughly five times and one-fifth of Pu-238, respectively. This conclusion was similarly arrived at by the European Space Agency (2012) when it decided to pursue the use of Am-241 as a fuel for its future RTG

space missions. A detailed analysis of an encapsulated Am_2O_3 fuel pellet is discussed further by Tinsley [6].

As previously mentioned, the most essential characteristics of a favorable nuclide include a large power density, a long half-life, and low radiation shielding requirements. Another important aspect worth noting, however, is the isotope's abundance in nuclear spent fuel; for the purposes of this project, reprocessing is considered the most viable method for the production of the discussed isotopes. Finally, it is beneficial if the daughter products of the isotope exhibit similar qualities. That is, that the decay product of the isotope satisfactorily meet all or some of the aforementioned criteria. Tabulated data for all of these features have been provided in Tables I and II below.

Table 2: Data for candidate Isotopes and their decay products' properties.[1]

	Pu-238	Am-241	Po-210	Cm-244	Cm-242	Sr-90
Half-Life	87.7 years	432.6 years	138.4 days	18.11 years	162.9 days	28.79 years
Power Density Average (Watts/Kilogram)	571	103	122,000	2,830	119,000	165
Class of Emitters	Alpha	Alpha	Alpha	Alpha	Alpha	Beta
Output from fuel cycle (percentage)	3.15E-05	2.70E-05	3.46E-20	1.44E-06	8.84E-07	3.11E-04
Primary Decay Product	U-234	Np-237	Pb-206	Pu-240	Pu-238	Y-90
Primary Decay Product Half-Life	2.455e5 years	2.144e6 years	Stable	6561 years	87.7 years	2.667 days
Primary Decay Product Power Density (W/kg)	0.179	0.0201	Stabe	7.07	562-584	3,008,000
Primary Decay Product Class Emitter	Alpha	Alpha	Stable	Alpha	Alpha	Beta
Generation 2 Decay Product	Th-230	Pa-233	None	U-236	U-234	Zr-90
Gen. 2 Decay Product Half-Life	75,381 years	26.975 days	N/A	2.34e7 years	2.455e5 years	Stable
Gen. 2 Decay Product Power Density (W/kg)	0.582	52740	N/A	0.002	0.179	Stable
Gen. 2 Decay Product Class Emitter	Alpha	Beta	N/A	Alpha	Alpha	Stable

[1] *Note that an asterisk denotes the isotope is metastable.*

Table 2 (Continued): Data for candidate isotopes and their decay products' properties.

	Cf-252	Ce-144	Cs-137	Pm-147	Ru-106	Co-60
Half-Life	2.645 years	284.91 days	30.08 years	2.62 years	1.018 years	5.271 years
Power Density Average (Watts/Kilogram)	38,100	2260	97	340	197	18,900
Class of Emitters	Alpha + SF 3.1%	Beta	Beta	Beta	Beta	Beta
Output from fuel cycle (percentage)	7.28E-18	2.10E-04	6.39E-04	1.35E-04	6.45E-05	0.00
Primary Decay Product	Cm-248	Pr-144 or Pr*-144	Ba*-137 or Ba-137	Sm-147	Rh-106	Ni-60
Primary Decay Product Half-Life	3.480e5 years	17.28 minutes or 7.2 minutes*	2.552 minutes* or Stable	1.06e11 years	30.07 seconds	Stable
Primary Decay Product Power Density (W/kg)	0.535	5.55e8 or 6.236e7*	2.114e9* or Stable	3.14E-07	3.43E+10	Stable
Primary Decay Product Class Emitter	Alpha + SF 8.4%	Beta or Isomeric Transition*	Isomeric Transition* or Stable	Alpha	Beta	Stable
Generation 2 Decay Product	Pu-244	Nd-144	None	Nd-143	Pd-106	None
Gen. 2 Decay Product Half-Life	8.11e7 years	2.29e15 years	N/A	Stable	Stable	N/A
Gen. 2 Decay Product Power Density (W/kg)	5.31E-04	1.34E-11	N/A	Stable	Stable	N/A
Gen. 2 Decay Product Class Emitter	Alpha + SF 0.125%	Alpha	N/A	Stable	Stable	N/A

Conclusions

As iterated in the results, Am-241 seems to be the candidate which exhibits the most potential for long term missions. While it does yield a higher percentage of the spent fuel examined than many other candidate isotopes, there are already existing methods of production for Am-241 that could make its implementation much more feasible. Another promising candidate is Sr-90. The availability of Sr-90 in spent nuclear fuel is favorable (~3.11E-04 percent) and the power density it provides is slightly higher than that of Am-241. The major limitation with Sr-90 is its relatively short half-life (~29 years), however this may not be an issue if it is implemented on shorter projects within the solar system. An isotope of particular interest is Cm-242, primarily due to its alpha decay into Pu-238 with a half-life of roughly 0.45 years and a power density of 119 kW/kg. The possibility of using a decay chain to power RTGs is worth exploring in future research and could prove very useful in forthcoming long-term space missions.

One unfortunate feature of the results is that most of the isotopes considered have a lower yield in spent fuel than Pu-238. This is to be expected when considering the transmutation of nuclear fuel, shown in Fig. 2.

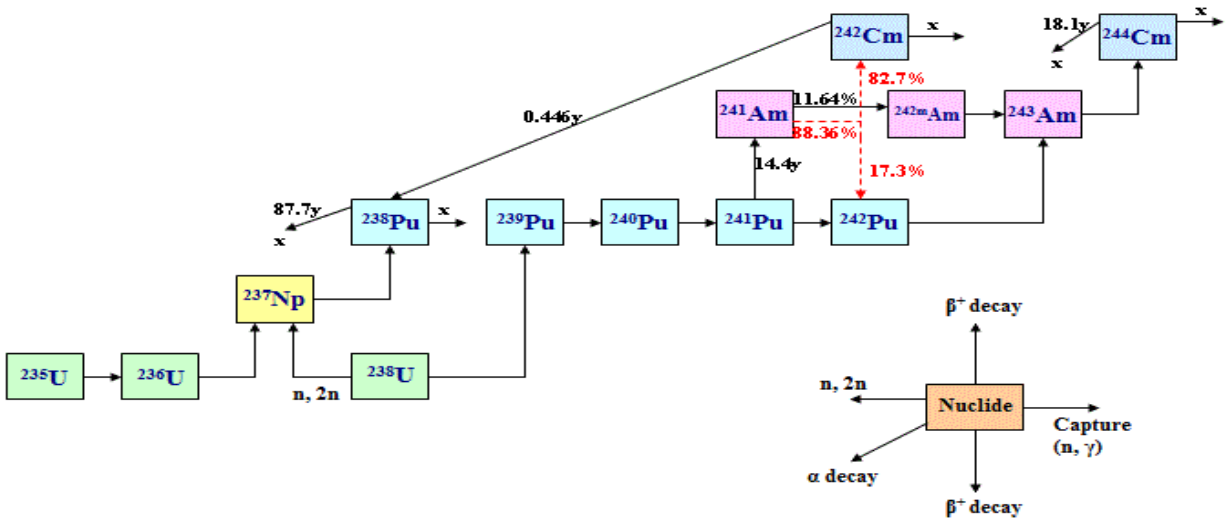

Figure 6: Transmutation in Nuclear Fuel compositions [7]

It should again be noted that a major limitation to this project is the fact that many of the isotopes considered are not feasibly extracted from spent nuclear fuel with current isotope separation methods and facilities, and that many times it is much more economical to produce the prospective isotopes through irradiation production. However, it is difficult to ignore the underlying value of spent nuclear fuel beyond recycling for the power production cycle.

Although there are promising candidates to replace Pu-238, further investigation into viable fuel alternatives for RTGs is undoubtedly necessary if deep space is ever to be explored.

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