OPTIMIZATION OF PLUTONIUM-238 PRODUCTION IN THE ADVANCED TEST REACTOR FOR RADIOISOTOPE THERMOELECTRIC GENERATORS IN DEEP SPACE EXPLORATION APPLICATIONS

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Plutonium production for use in Radioisotope Thermoelectric Generators (RTGs) is becoming increasingly important in the United States due to several reasons. The plutonium stockpile originally purchased from the USSR has steadily decreased due to utilization in RTGs and the degrading quality of the specific plutonium isotope of interest in the stockpile: plutonium-238. With a half-life of 87.7 years, the quality has decreased since it was originally purchased just after the Cold War. The United States is aiming to produce high-quality ^{238}Pu and become independent in space fuel production in the process. The high-quality plutonium combined with the currently available low-quality plutonium will produce usable fuel for future space explorations like the Mars 2020 expedition. This paper details the new models / methodologies investigated by Center for Space Nuclear Research (CSNR) to optimize ^{238}Pu production in the Advanced Test Reactor (ATR). Increasing the ^{238}Pu production at ATR will supplement the current production at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Labs in Tennessee to achieve the national goal of 1.5 – 2.0 kg ^{238}Pu per year.

I. PROJECT BACKGROUND AND SPECIFICATIONS

I.A. Plutonium-238

The space industry is particularly interested in ^{238}Pu for assorted reasons:

- alpha particle emitter – provides high heat output,
- favorable half-life – the energy released is at a constant rate for a reasonable amount of time,
- easily produced – exist in insoluble form, high melting point,
- relatively safe to handle – sintered ceramic pellets minimize contamination,
- requires little shielding – alpha particles can be blocked with as little as a piece of paper, and
- high-power density – fuel must produce substantial amount of power per mass and volume.

Figure 1 shows the reaction schemes available for transmuting neptunium into plutonium.

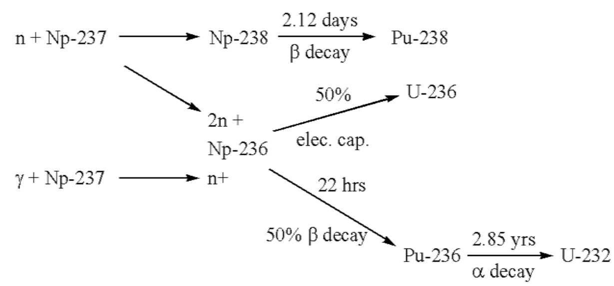


Fig. 1. Reaction schemes for transmuting Np into Pu (Credit: Patent US 6896716 B1)

As shown in Figure 1, the desired reaction is an (n, γ) reaction with ^{237}Np that results in ^{238}Np. The most unfavorable reaction in an (n, 2n) reaction that results in the buildup of ^{236}Pu. This isotope is highly radioactive with deeply penetrating gamma rays. For these reasons, the assay of the final product is dependent on the buildup of ^{236}Pu. Fortunately, the (n, 2n) reaction occurs primarily with high energy neutrons above 0.6 MeV which is in the “fast neutron” energy range. Therefore, the unfavorable reaction can be minimized with proper neutron moderation.

I.B. Radioisotope Thermoelectric Generators (RTG)

An RTG is an electrical generator that uses an array of thermocouples to convert thermal energy (heat released by the spontaneous decay of a radioactive material) to electrical energy using the Seebeck effect. RTGs are used in deep space spacecrafts where it is impractical to use any other form of energy. ^{238}Pu is an alpha emitter that produces a significant amount of heat. The half-life of

^{238}Pu (87.7 years) is beneficial because the release of alpha particles is fast enough to produce usable heat but slow enough to power equipment for 80-160 years. This enables the spacecraft to travel out of the solar system without refueling.

Figure 2 shows a system of diagrams that identify where the plutonium fuel pellet is placed in a general-purpose heat source, which is then used in the assembly of a complete RTG.

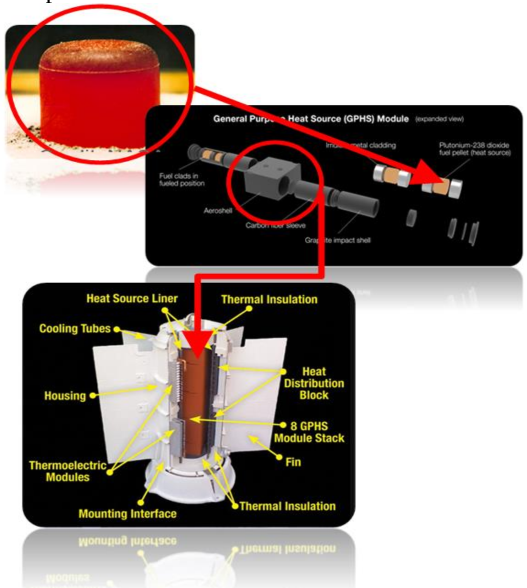


Fig. 2. Plutonium fuel pellet, general purpose heat source diagram, and RTG diagram. (Credit: NASA)

I.C. Advanced Test Reactor

The Advanced Test Reactor is located in the ATR complex of the Idaho National Laboratory site. ATR is rated at a peak power of 250 MW_{th} and has been in continuous operation since 1967. There are irradiation locations in the outer rim of the reactor that are of less demand and are an ideal location for ^{238}Pu production. These are the I-positions represented by the color red. The primary reason for potential use of the I-positions is due to the high thermal neutron flux that is ideal for a high assay ^{238}Pu yield. These positions also have low fast neutrons, so ^{236}Pu production should not be large. These areas are shown in Figure 3, which shows a top-down cross section cutaway of the ATR core.

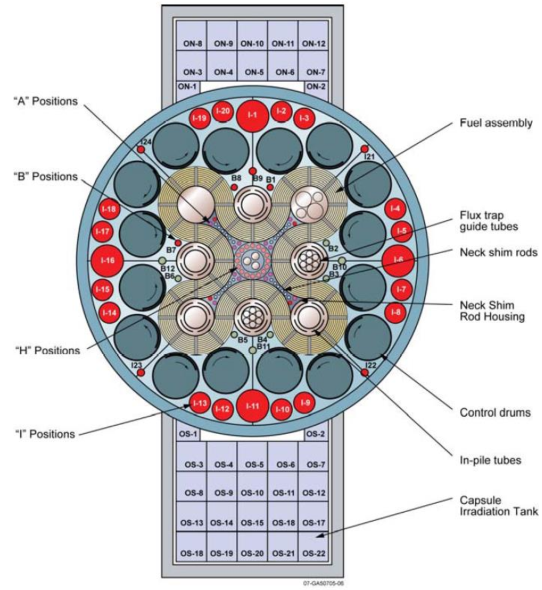


Fig. 3. Top-down cross section of the ATR (Credit: Advanced Test Reactor National Scientific User Facility User's Guide)

II. OPTIMIZATION

MCNP and Serpent Modeling Results

The ^{238}Pu and impurity concentrations were modeled using MCNP as well as the Monte-Carlo code Serpent. Serpent is very similar to MCNP in many regards, especially in the way inputs are specified and k_{eff} is estimated, but it includes more modern features which MCNP lacks. Namely, isotope tracking. This feature has been built in to the code from its inception and is quite sophisticated. MCNP is the gold standard for reactor modeling, while Serpent is still relatively new to the nuclear community.

Using the 94CIC (1994 Core Internals Changeout) benchmark model of the ATR (the same model that was used in MCNP, but re-written in Serpent), Np pellets were placed in various I-positions at 105 MW_{th},, and all the relevant isotopes were tracked. The main isotopes of concern were Plutonium 236 and Plutonium 238. The Plutonium 236 isotope is important since it presents a radiological hazard to workers, and its concentration must be kept to a minimum. By default, Serpent uses the ENDF VII.I cross sections, which are reliable for most of the isotopes of concern but not for this case.

One type of pellet was tested. The makeup was 20% NpO_2 and 80% Al. In all cases, the pellet stacks were approximately 19 inches tall, and were centered at the center of the reactor. The number and mass densities were computed for numerous Np and Pu isotopes. Pellets were tested in four different configurations.

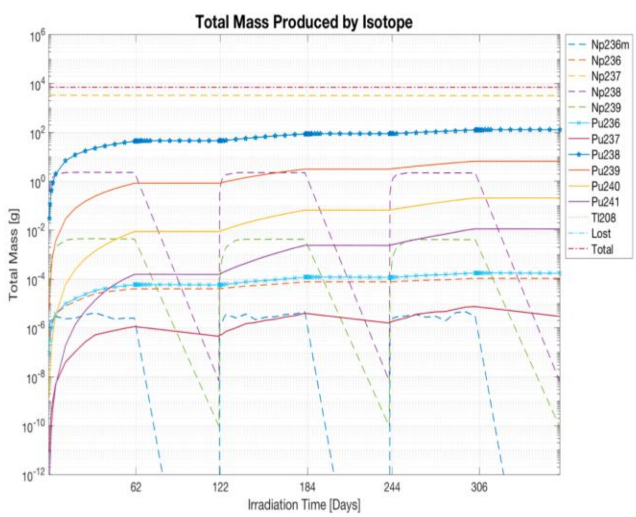


Fig. 4. Serpent comparison of total mass produced of each isotope

Expected values of ^{236}Pu were to be in the parts per million range. The results shown in Figure 5 display that the amount of ^{236}Pu produced in the Serpent results agree with the expectations given.

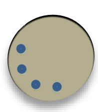
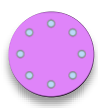
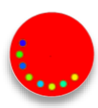
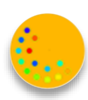
Design	**#1**	**#2**	**#3**	**#4**
				
^{238}Pu Mass [g]	129	204	193	267
Conversion Rate [%]	3.83	3.03	3.25	2.46
^{236}Pu [ppm] [Mg/g]	1.17	1.08	1.28	1.65
^{239}Pu mass [g]	6.68	8.24	8.40	8.60
^{238}Pu Quality	95%	96%	96%	97%

Fig. 5. Serpent Design Comparisons

All four designs fell into the expected range of ^{236}Pu produced. The relationship between ^{238}Pu produced and the number of rods can be seen as well. An increase in the number of rods leads to an increase in the amount of ^{238}Pu produced. This was a reasonable expectation. The placement of the rods also dictates the efficiency of ^{238}Pu production. The goal of working with designs 2 and 3 were to see how the location of the rods affect the production of ^{238}Pu. It was found that the locations of the rods do have a noticeable impact on the amount of ^{238}Pu produced.

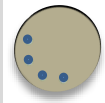
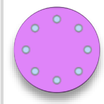
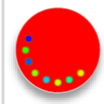
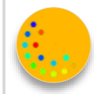
Design	**#1**	**#2**	**#3**	**#4**
				
^{238}Pu Mass [g]	147	231	222	331
Conversion Rate [%]	4.37	3.44	4.04	3.05
^{236}Pu [ppm] [Mg/g]	Not Calc.	Not Calc.	Not Calc.	Not Calc.
^{239}Pu mass [g]	5.717	8.33	7.25	8.97
^{238}Pu Quality	96%	96%	97%	97%

Fig. 6. MCNP Design Comparisons

The MCNP calculations were done to check the validity of the Serpent results. As can be seen from comparing Figures 5 and 6, the MCNP results are in close relation to the Serpent results.

Design	**#1**	**#2**	**#3**	**#4**
Efficiency	0.038	0.030	0.033	0.025
Total Pu 238 (g)	129	204	193	267
Quality	0.95	0.96	0.96	0.97
Pu 236 (ppm)	1.17	1.08	1.28	1.65
Number of rods	4	8	8	14
Analysis Factor	**0.9963**	**0.6842**	**0.5903**	**0.2760**

Fig. 7. Unweighted Design Comparison (Red indicates a negative quality factor)

An unweighted design comparison was developed to give a general idea on which design would be best based on which quality is valued most. The overall final value was calculated by multiplying each positive quality factor and dividing by each negative quality factor (shown in red). A cost factor could not be produced because there was no literature on the amount each individual rod would cost to produce. The number of rods factor was used to give a general idea on the cost of each design. For an unweighted factor, design 1 had the best analysis factor. The transparent decrease in the analysis factor from design 1 to 4 is probably due to the number of rods. For future analysis work, this should be weighted to be of less importance. It seems to have the strongest effect on the design comparison as of now.

Design	85% (g)
#1	280
#2	464
#3	440
#4	634

Fig. 8. Estimated ^{238}Pu Production from Serpent using the current stockpile of low quality ^{238}Pu

Currently, the US has a stockpile of low-quality plutonium, around 78%, due to decay. The use of the high-quality plutonium from the ATR can be mixed with the stocklpile to increase the enrichment to 85% ^{238}Pu. This will increase the amount of usable plutonium produced per year. As seen in Figure 8, the inclusion of the old plutonium increases the yearly yield of the ATR by almost a factor of 2.

III. CONCLUSIONS

Using Serpent, results were found on the amount of ^{238}Pu that could be made per year using the Advanced Test Reactor I-positions. On average, approximately 200 grams of ^{238}Pu could be produced per year. NASA is interested in producing 1.5-2 kilograms of ^{238}Pu per year for future space missions. This means realistically, NASA could send out one probe every 10 years using the highly enriched ^{238}Pu produced from the ATR. By enriching the stockpile to 85%, the usable ^{238}Pu can be doubled. This would allow NASA to send a probe every 5 years. The High Flux Isotope Reactor (HFIR) located at Oak Ridge National Lab is already producing ^{238}Pu, but the produced amount is highly dependent on the demand of the HFIR's outer positions.

Serpent was used to model four different designs, each varying the number of rods and locations of rods in the I-positions. MCNP was used to model all four designs as well to check the validity of Serpent. The results from MCNP and Serpent were in close agreement.

Each design presented different advantages and disadvantages. An overall unweighted analysis factor was able to be found for each design. The factor seemed highly reliant on the number of rods, therefore future analysis should include weighted factors.

IV. FUTURE WORK

Placing targets in the ATR and getting experimental results would be useful to check the computational results from this project. Thermal limitations were not considered and should to be taken into account for practical use of the optimal design. As seen from design 1 and 2, the location of the rods affects the efficiency of production. More research should be done to find the optimal geometries which will produce the highest efficiency of production in the I-positions.

ACKNOWLEDGMENTS

We wish to thank Dr. Herring from CSNR, Brian Gross, Doug Crawford, and Mark DeHart from INL, Dr. Paul Wilson from the University of Wisconsin, and Jakko Leppänen from VTT for their support of this project.

This research made use of the resources of the High-Performance Computing Center at Idaho National Laboratory which is supported by the Office of Nuclear Energy of the U.S. Department of Energy and the Nuclear Science User Facilities under Contract No. DEAC0705ID14517.

In addition, this research used ENDF VII.I cross section data for calculations from JANIS 4.0.

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