INL/CON-22-65857-Revision-0

Neutronic Safety Analysis of Pu-238 Production at Idaho National Laboratory

May 2022

Changing the World's Energy Future

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Idaho National Laboratory

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Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

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This analysis was completed to support the irradiation of plutonium-238 production targets in the North East Flux Trap (NEFT) in the Advanced Test Reactor (ATR) as a part of the campaign to restart domestic production of plutonium-238 used in radioisotope power systems (RPS) by the National Aeronautical and Space Administration (NASA) and Department of Energy (DOE) Office of Nuclear Energy (NE), Office of Nuclear Infrastructure Program (NE-3). Referred to as the PFS-ATR-GEN1-NEFT experiment, the assembly was designed to hold 46 Pu-238 production targets in the NEFT. The scope of this paper is to outline the MCNP to ORIGEN2 in Python (*MOPY) method used to calculate the heat generation rates, flux, and fission density and to quantify the viability of the target design for Pu-238 production in ATR.*

I. INTRODUCTION

This analysis was completed to support the irradiation of plutonium-238 production targets in the North East Flux Trap (NEFT) in the Advanced Test Reactor (ATR) as a part of the campaign to restart domestic production of plutonium-238 used in radioisotope power systems (RPS) by the National Aeronautical and Space Administration (NASA) and Department of Energy (DOE) Office of Nuclear Energy (NE), Office of Nuclear Infrastructure Program (NE-3).

Pu-238 production at Idaho National Laboratory (INL) will use Oak Ridge National Laboratory manufactured targets, which are referred to as ATR Generation I Targets. Each Pu-238 production target consists of a stack of cylindrical pellets, composed of 20-volume% neptunium oxide (NpO_2), 70-volume% aluminum, and 10-volume% void as well as an aluminum / samarium spacer pellet on the top of each stack.

Referred to as the PFS-ATR-GEN1-NEFT experiment, the total assembly was designed to hold 46 Pu-238 production targets in the NEFT, two targets in each NEFT position with the neptunium pellet material axially centered about core midplane, the upper target will be rotated so that the tops of the two targets are nose to nose with the Al/Sm spacers near the core mid-plane, see Figure 1.

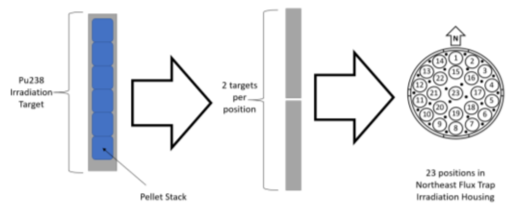

Fig. 1. PFS-ATR-GEN1_NEFT Assembly.

The scope of this paper is to outline the MOPY method used to calculate the heat generation rates (HGRs), flux, and fission density and to quantify the viability of the target design for Pu-238 production in ATR.

II. MOPY METHODOLOGY

The particle transport (neutron and photon) analyses were completed using MCNP, a general-purpose Monte Carlo N-Particle transport code. The output produced by MCNP was then used to complete the activation and depletion analysis in ORIGEN2.

INL developed a python-based code coupling MCNP and ORIGEN2, called MOPY (MCNP to ORIGEN2 in Python). MOPY was written using the python scripting language and was developed to manage input file generation, execute MCNP5, extract and manipulate the MCNP output, transfer the data into the ORIGEN2 input file, and execute ORIGEN2.

II.A. Purpose of MOPY

The purpose of MOPY is (1) to calculate the depletion/irradiation analysis of materials in ATR. (2) Have a standardized method available to analyze materials in ATR. (3) Calculate the following: (3i) actinide, fission product, and/or activation product inventories as a function of irradiation parameters (time, flux, operating conditions, material location in reactor); (3ii) neutron and photon heat generation rates (fuel and non-fuel materials); (3iii) radionuclide isotopic source terms; (3iv) radionuclide decay heat source terms; (3v) and photon spectrum for photon source terms.

II.B. MOPY Assumptions

To use MOPY it should be understood that: (1) The user has licenses to MCNP, ORIGEN2, and data libraries. (2) The user has an error-free MCNP5 "base" model. To designate the cells of interest, keyword phrases are used to delimit blocks for cell and material cards. (3) The user creates unique MCNP material cards to define the tracked al ORIGEN2 calculations. (4) MCNP reactions used to generate substitute cross-sections for each tracked nuclide for all cells/materials being irradiated, depleted, or activated are (a) actinides - (n, γ), (n, 2n), (n,3n), (n, fission); with MCNP reaction numbers of 102, 16, 17, -6; (b) fission products and light elements -(n, γ), (n, 2n), (n,3n), (n, proton); with MCNP reaction numbers of 102, 16, 107, and 103. (5) The MCNP-calculated ORIGEN2 one-group substitute cross-sections for each tracked nuclide for each reaction type tracked for all depleted cells/materials are: (a) ORIGEN2 one-group cross-section IDs for actinides - SNG, SN2N, SN3N, SNF (b) ORIGEN2 one-group cross-section IDs for fission products and light elements - SNG, SN2N, SNA, SNP. (6) An MCNP material composition irradiated, depleted, or activated may not have duplicate nuclides with different cross section IDs defined (e.g. 13027.70c and 13027.37c cannot both be defined for a material composition). (7) MCNP material compositions irradiated, depleted, or activated are defined using atom densities. (8) ORIGEN2 material compositions irradiated, depleted, or activated are defined using gram-atoms (moles). (9) Material compositions used for the ORIGEN2 calculations are defined using a volumetric basis. One cubic centimeter (1 cc) of the material composition from MCNP is used to define the ORIGEN2 material composition (gram-atoms/cc for ORIGEN2 and atoms/barn-cm for MCNP; use Avogadro's number for conversion between these values). This assumption can be verified by comparing the total mass reported in the ORIGEN2 output with the density used.

II.B. Running MOPY

To run MOPY the input file was created specifying the specific ATR cycle data. The materials and reaction rates of interest were also captured in the input file. The input file was then executed and the updated MCNP file ran. Once the MCNP run was completed the flux and appropriate material cross-sections were fed into ORIGEN2 and ORIGEN2 was executed. This cycle was repeated for the desired number of irradiation days.

To complete the Pu-238 production target analysis, MOPY was ran for 9 timesteps to capture 65 days of irradiation. Each timestep varied from 3-10 days in length but all assumed a constant lobe power of 20 MW. For the Pu-238 production target analysis approximately 70 nuclides were tracked in MOPY.

The ENDF/B-VII.0 cross section library that comes with MCNP was used along with the neptunium-236m cross section library obtained from TENDL-2017. The standard ATR cross section library was used for ORIGEN2 along with MCNP-calculated replacement cross sections. While multiple nuclides were tracked in MOPY the tracking of Np-236m (See Fig. 2) was important for tracking production of the impurity Pu-236 and enabling its minimization due to significant gamma emitters in its decay chain. To properly track the amount of Np-236m produced from the (n,2n*) reaction, where the * indicates an excited or metastable state, MOPY must be properly set to handle this reaction. It has three different treatment settings (default, zero, and ratio) for calculating the cross section that leaves a product nucleus in a metastable state, and the ratio setting was deemed most prudent for this analysis. In the ratio setting, MOPY calculates the ratio of the (n,2n) reaction cross section to the (n,2n*) cross section in the standard cross section library and then applies that ratio to the MCNP-calculated (n,2n) total cross section to the calculate the new (n,2n) and (n,2n*) replacement cross sections used by ORIGEN.

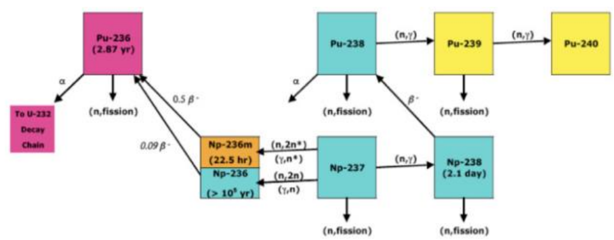

Fig. 2. Nuclear Reactions of Interest for Neptunium Pellet Material.

III. MODEL DESIGN

Each Pu-238 production target consists of a stack of cylindrical pellets, composed of 20-volume% neptunium oxide (NpO_2), 70-volume% aluminum, and 10-volume% void. Two targets are stacked end-to-end and placed into a basket. This design was chosen to increase the number of NpO_2-Al pellets irradiated per cycle and to utilize the full length of the ATR, see Figure 1.

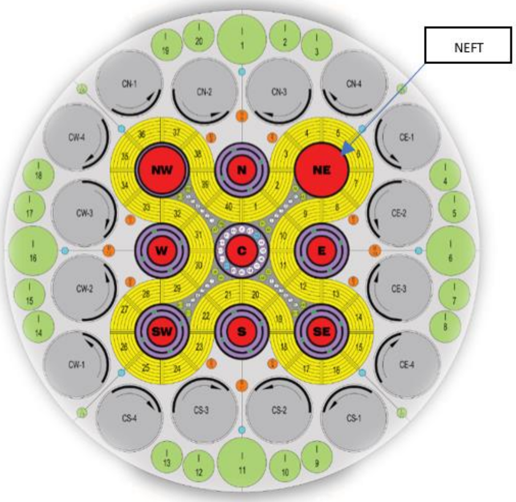

Fig. 1. Cross section of the ATR core.

The following is a summary of the structural hardware used in the target assembly. An aluminum 6061-T6 and samarium tubular spacer assembly are placed at the lower end of the pellet train and an Al-4047 autogenous weld backing tube is installed at the top of the pellet train. The target cladding consists of finned Aluminum 6061-T4 cladding extrusion that contains the pellets and spacers, that are welded to Al 6061-T6/stainless steel 304L bimetallic transition joint parts that make up the top and bottom terminations of the target. A stainless-steel cap is installed in the bottom of the target to seal the target. For purposes of the MCNP analysis most of the structural hardware was homogenized together. This homogenization has a minimal impact on the neutronic analysis.

IV. CALCULATIONS

By utilizing MCNP and ORIGEN2, via MOPY, multiple analyses were completed for the beginning of cycle and end of cycle conditions. The following sub-sections outline the methods used to complete the analyses.

IV.A. Core Power Calculation

Each corner lobe in ATR is designed to operate individually, the core power used in calculating heat generation rates and flux must be scaled to the nearest lobe. This is done in the following fashion:

By tallying the fission energy in the driver fuel in each lobe, summing them, the core fission energy is calculated as is shown in Equation (1). The lobe power is then calculated in Equation (2) by multiplying the expected core power by the lobe energy fraction of the calculated core energy. The expected core power is the sum of all the lobe powers for a given ATR Cycle. The scaled core power is then calculated by dividing the expected lobe power by the calculated lobe and multiplying by the expected core power as shown in Equation (3).

$$\begin{aligned} &\mathit{Calculated\ Core\ Energy}\ (MeV) \\ &= \sum_i^4 \left[\mathit{Calculated\ Lobe\ Fuel\ Mass}\ (g)_i \right. \\ &\left. \times \mathit{Calculated\ Lobe\ Fuel\ F7\ Tally} \left(\frac{MeV}{g}\right)_i \right] \end{aligned} \quad (1)$$

$$\begin{aligned} &\mathit{Calculated\ Lobe\ Power}\ (MW) \\ &= \mathit{Expected\ Core\ Power}\ (MW) \\ &\times \frac{\mathit{Calculated\ Lobe\ Energy}\ (MeV)}{\mathit{Calculated\ Core\ Energy}\ (MeV)} \end{aligned} \quad (2)$$

$$\begin{aligned} &\mathit{Scaled\ Core\ Power}(MW) \\ &= \mathit{Expected\ Core\ Power}\ (MW) \\ &\times \frac{\mathit{Expected\ Lobe\ Power}\ (MW)}{\mathit{Calculated\ Lobe\ Power}\ (MW)} \end{aligned} \quad (3)$$

IV.B. Heat Generation Rates

MCNP reports tally results normalized per source particle. The MCNP type 6 energy deposition tally results or type 7 fission energy deposition tally results are used to calculate heat generation rates. The MCNP tally type 6 has units of MeV/g per source particle (fission neutron for prompt neutron, gamma heating, and fission heating.

The heat generation rate values are calculated using the MCNP tally type 6 results, the heating normalization factor (HNF), and the ATR core power. Prompt neutron and gamma heating rates (PHR) are calculated using equation (5).

$$\mathrm{PHR}=(\mathrm{f6})(\mathrm{HNF})(\mathrm{Core\ Power})\ \frac{W}{g} \quad (5)$$

The linear heat generation rates are calculated by multiplying the PHR (see equation 5) by the MCNP calculated volume and then dividing that product by the segment (in cm). The power density was calculated by multiplying the PHR by the MCNP calculated mass and then dividing that product by the MCNP calculated volume (cc) for that segment.

IV.C. Fission Density Calculation

The fission density was calculated using the output files produced from MOPY. Fission density is a cumulative measure of fissions that have occurred per unit volume in fissionable material and is directly related to burnup. Because each fission typically produces fission fragments that have atomic numbers, Z, less than 90 it can be assumed that every fission removes a heavy metal atom (Z≥90) from the fissionable material. Thus, the deficit between the initial density of heavy metal, $\rho_{initial}$, atoms and the current density of heavy metal atoms, $\rho_{current}$, is how the fission density, FD, is calculated.

V. RESULTS

V.A. Heat Generation Rates

The HGRs for the PFS-ATR-Gen1 experiment were calculated using a 3 radial, 7 axial-region fuel model of ATR. The peak HGRs in the neptunium pellet material are reported in the following table (Table I). The peak fission density in the neptunium pellet material in fissions/cc at each timestep are also reported. Figure 3 shows the heating profile of the Pu-238 production targets. The peak heating occurred in position 23 of the NEFT.

TABLE I. Np Pellet Heating Summary.

EFPDs	Peak f6 Tally (neutron + photon + delayed photon) HGR (W/g)	Peak Fission Density (fission/cc)
0	26.60	0.00
65	326.40	1.93E+20

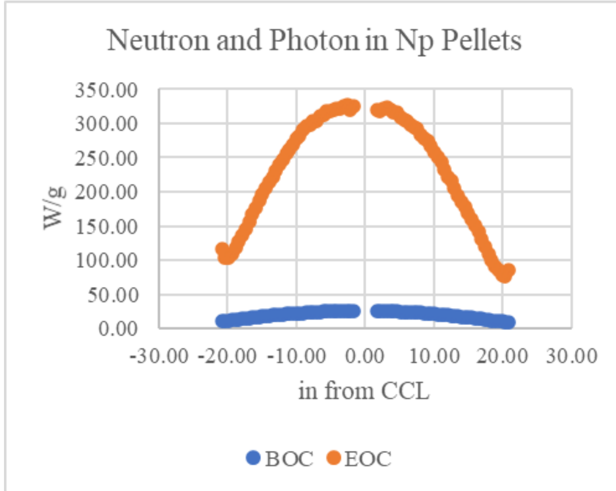

Fig. 3. Heating profile of Pu-238 production targets.

V.B. Pu-238 Yield

The following table (Table II) reports the average fission density, average Pu-238 Assay, average Np Conversion, average Pu-236 Content, and the Pu-238 Yield per target. This information is reported after 60 days of irradiation as this is the nominal ATR cycle length.

TABLE II. Estimated Peak Pu-238 Yield for the NEFT.

Position	Average Fission Density per target	Average Pu-238 Assay per target	Estimated Peak Pu-238 Yield (g) per target*
Upper Tube 1	6.15E+19	88.39%	2.82
Lower Tube 1	6.58E+19	87.98%	2.92
Upper Tube 2	6.00E+19	88.49%	2.80
Lower Tube 2	6.39E+19	88.11%	2.89
Upper Tube 3	5.97E+19	88.53%	2.79
Lower Tube 3	6.38E+19	88.12%	2.88
Upper Tube 4	6.20E+19	88.35%	2.83
Lower Tube 4	6.73E+19	87.86%	2.87
Upper Tube 5	6.61E+19	88.07%	2.90
Lower Tube 5	7.13E+19	87.61%	3.01
Upper Tube 6	7.18E+19	87.71%	2.99
Lower Tube 6	7.78E+19	87.19%	3.11
Upper Tube 7	7.87E+19	87.26%	3.08
Lower Tube 7	8.59E+19	86.70%	3.22
Upper Tube 8	8.34E+19	86.82%	3.17
Lower Tube 8	9.16E+19	86.21%	3.31
Upper Tube 9	8.65E+19	86.60%	3.21
Lower Tube 9	9.55E+19	85.94%	3.36
Upper Tube 10	8.36E+19	86.78%	3.18

Position	Average Fission Density per target	Average Pu-238 Assay per target	Estimated Peak Pu-238 Yield (g) per target*
Lower Tube 10	9.25E+19	86.14%	3.33
Upper Tube 11	8.23E+19	86.86%	3.16
Lower Tube 11	9.06E+19	86.23%	3.30
Upper Tube 12	7.59E+19	87.31%	3.07
Lower Tube 12	8.34E+19	86.72%	3.21
Upper Tube 13	6.93E+19	87.76%	2.98
Lower Tube 13	7.57E+19	87.21%	3.10
Upper Tube 14	6.42E+19	88.11%	2.90
Lower Tube 14	6.95E+19	87.63%	3.01
Upper Tube 15	8.34E+19	86.20%	3.11
Lower Tube 15	8.98E+19	85.71%	3.21
Upper Tube 16	8.17E+19	86.31%	3.09
Lower Tube 16	8.78E+19	85.82%	3.19
Upper Tube 17	8.51E+19	86.11%	3.13
Lower Tube 17	9.19E+19	85.61%	3.24
Upper Tube 18	9.25E+19	85.71%	3.22
Lower Tube 18	1.01E+20	85.13%	3.34
Upper Tube 19	9.92E+19	85.32%	3.29
Lower Tube 19	1.09E+20	84.70%	3.43
Upper Tube 20	1.01E+20	85.24%	3.31
Lower Tube 20	1.11E+20	84.58%	3.45
Upper Tube 21	9.68E+19	85.46%	3.27
Lower Tube 21	1.06E+20	84.83%	3.40
Upper Tube 22	8.94E+19	85.87%	3.19
Lower Tube 22	9.72E+19	85.30%	3.30
Upper Tube 23	1.11E+20	84.33%	3.36
Lower Tube 23	1.20E+20	83.72%	3.48
Max	1.20E+20	88.53%	3.48
Min	5.97E+19	83.72%	2.79
		Total:	144.41

* actual production values may vary

VI. CONCLUSIONS

Pu-238 production at Idaho National Laboratory will be completed using Oak Ridge National Laboratory manufactured targets, which are referred to as PFS ATR Generation I Targets. To support irradiation of the PFS ATR GEN-1 targets multiple analysis were completed using the python-based code, MCNP to ORIGEN2 in Python (MOPY). MOPY was written using the python scripting language and was developed to manage input file generation, execute MCNP5, extract and manipulate the MCNP output, transfer the data into the ORIGEN2 input file, and execute ORIGEN2.

This report outlined the MOPY method used to calculate the heat generation rates (HGRs), fission density, and quantify the viability of the target design for Pu-238 production in ATR.

If the Pu-238 production experiment is irradiated for 65 days in the NEFT at a NE lobe power of 20 MW, the neptunium pellet material undergoes a rapid increase in heating in the first few days followed by a more gradual increase in heating until the end of the cycle.

The maximum heat generation rate is 326.40 W/g (peak f6 tally) with an associated power density of 1333.58 W/cc. The maximum fission density is 1.93E+20 fissions/cc when the fission density is averaged over each target. These values are acceptable for design and irradiation.

It is estimated that irradiating 46 PFS ATR GEN-1 targets in the ATR NEFT for 60 days will produce a maximum amount of 144g of Pu-238. The qualification and production of Pu-238 in the NEFT of ATR puts INL on track to meet or exceed the Pu production goals by 2025.

ACKNOWLEDGMENTS

This work was funded through DOE & NASA Interagency Agreement # NNH19OB05A and DOE contract DE-AC05-00OR22725 and DE-AC07-05ID14517. This research also 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. DE-AC07-05ID14517.

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