INL/JOU-23-75073-Revision-0

Progress on Pu-238 Production at Idaho National Laboratory from February 2022 to July 2023.

September 2024

Changing the World's Energy Future

William Spencer Green, Andrew John Zillmer, Justin D Lower, Jill R Mitchell, Brittany Jean Grayson, Erik S Rosvall, Austen David Fradeneck, Joshua Fishler, Ryan L Marlow, Mark A Hill

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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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CASE STUDY

Progress on Pu-238 production at Idaho National Laboratory from February 2022 to July 2023 [version 1; peer review: 1 approved]

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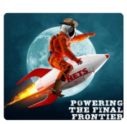
v1 **First published:** 19 Sep 2024, **2**:66 https://doi.org/10.12688/nuclscitechnolopenres.17503.1
Latest published: 19 Sep 2024, **2**:66 https://doi.org/10.12688/nuclscitechnolopenres.17503.1

Abstract
Idaho National Laboratory (INL) has continued to qualify irradiation positions in the Advanced Test Reactor (ATR) for Pu-238 production to support NASA deep space missions. Over the past year, INL qualified Np-237 targets for ATR's North East Flux Trap (NEFT), Inner-A and H positions. Work has begun to requalify the South Flux Trap (SFT) and to qualify the East Flux Trap (EFT) for the ATR GEN I target and is midway through the qualification process. This paper gives an overview of operational and technical activities from February 2022 to July 2023.

Keywords
Pu-238, Plutonium, Pu-238 Production, Plutonium Production



This article is included in the Research from the Nuclear and Emerging Technologies for Space (NETS) Conferences collection.

Open Peer Review

Approval Status ✓

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version 1 19 Sep 2024	✓ view

1. **Qingquan Pan**, Shanghai Jiao Tong University, Shanghai, China

Any reports and responses or comments on the article can be found at the end of the article.

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Author roles: Zillmer A: Conceptualization, Project Administration, Supervision, Writing – Original Draft Preparation, Writing – Review & Editing; **Green W**: Conceptualization, Project Administration, Supervision, Writing – Original Draft Preparation, Writing – Review & Editing; **Lower J**: Formal Analysis, Writing – Original Draft Preparation, Writing – Review & Editing; **Mitchell J**: Formal Analysis, Writing – Original Draft Preparation, Writing – Review & Editing; **Grayson B**: Formal Analysis, Writing – Original Draft Preparation, Writing – Review & Editing; **Rosvall E**: Formal Analysis, Writing – Original Draft Preparation, Writing – Review & Editing; **Fradeneck A**: Formal Analysis, Writing – Original Draft Preparation, Writing – Review & Editing; **Fishler J**: Formal Analysis, Writing – Original Draft Preparation, Writing – Review & Editing; **Marlow R**: Formal Analysis, Writing – Original Draft Preparation, Writing – Review & Editing; **Hill M**: Writing – Original Draft Preparation, Writing – Review & Editing

Competing interests: All authors are employees of Battell Energy Alliance and received a salary for this work.

Grant information: This work was funded by NASA Interagency Agreement No. NNH19OB05A and U.S. Department of Energy (DOE) Contract Nos. DE-AC05-00OR22725 and DE-AC07-05ID14517. This work also leveraged 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.
The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

How to cite this article: Zillmer A, Green W, Lower J *et al.* **Progress on Pu-238 production at Idaho National Laboratory from February 2022 to July 2023 [version 1; peer review: 1 approved]** Nuclear Science and Technology Open Research 2024, **2**:66 https://doi.org/10.12688/nuclscitechnolopenres.17503.1

First published: 19 Sep 2024, **2**:66 https://doi.org/10.12688/nuclscitechnolopenres.17503.1

I. Program overview

The purpose of this program is to contribute to the 1.5 kg per year, constant rate, production goal of Pu-238 in the United States by 2026, which is used as fuel in Radioisotope Power Systems that enable deep space NASA missions. To achieve this goal, the program has been working to qualify as many positions as possible for the insertion of the ATR GEN I. Currently the NEFT, Inner-A and H positions have been qualified for the insertion of the ATR GEN I target, and work is in progress to qualify the SFT and EFT. The SFT and I-7 positions have been qualified for the High-Flux Isotope Reactor (HFIR) GEN II design, described in Reference 1. The SFT is currently being requalified for the newer ATR GEN I targets.

The ATR GEN I target (Figure 1) design was implemented to utilize the full height of the ATR core while maintaining a common target to be used in both the ATR and HFIR. The design consists of stacking the targets, reflected around the ATR core centerline, in each position, which allows two targets to be used per position. A samarium pellet was also included at the nose of each target to reduce end effects. Utilizing the full height of the core will increase production by 40% to 50% as compared to a single target designed for the height of the HFIR reactor. This also allows the target to be processed at Oak Ridge National Laboratory (ORNL), whereas a target with the full height of the ATR core would not fit in the hot cells at ORNL.

II. Pu-238 production for upcoming cycles

II.A ATR Core Internal Changeout

ATR's Core Internals Changeout (CIC) refers to the changeout of reactor core components that are degraded by the high radiation environment. This degradation is caused by very high neutron radiation, beryllium/neutron reaction, thermal stresses, and normal wear and corrosion. For example, the beryllium/neutron reaction creates helium build-up leading to internal pressure that, over time, causes swelling and cracking in the beryllium reflector blocks. The degraded beryllium blocks can impede reactor operation due to stuck control rods, difficulty inserting/removing driver fuel, and possible fuel damage.

The CIC VI outage to replace ATR core internal components began on April 26, 2021, required approximately 11 months to complete or 332 days, and successfully ended on March 28, 2022. After completion of the CIC, nuclear testing was performed to ensure correct core operating parameters were initiated. Nuclear testing was successfully completed in November of 2022. It is expected that this CIC evolution will provide continued reactor operations for at least 10 years.

II.B Latest ATR cycle 171A irradiation

Cycle 171A was a 57-day cycle that started on April 26, 2023, and terminated on June 19, 2023. This was the first cycle where the newly designed ATR GEN I targets were inserted into the ATR. Due to target availability, INL only inserted 57 targets in the ATR for this cycle. Of the 57 targets, 46 ATR GEN I targets were inserted in the NEFT, two ATR GEN I targets in the inner-A position, two ATR GEN I targets in the H position, and seven HFIR GEN II targets in the SFT.

II.C Current cycle 171B

Cycle 171B is a projected 65-day cycle that was recently added to the ATR schedule and began on July 21, 2023. With the addition of this cycle, there were approximately 50 positions available which could accommodate 100 targets. However, due to target availability, only 49 targets were inserted into ATR. Of the 49 targets, 46 ATR GEN I targets were inserted in the NEFT and 2 in an H position. The final target was a spare HFIR GEN II target that was inserted in the SFT. Upon completion of this cycle, an estimated 210 g of heat source material will be produced.

II.D Upcoming cycle 172A

Cycle 172A is a 7-day operating cycle instead of the typical 60-day cycle. Therefore, targets will not be inserted in the cycle because the short cycle length would result in a low production amount of Pu-238.

II.E Upcoming cycle 173A irradiation

Cycle 173A is a 60-day cycle estimated to start in spring 2024. Analysis to increase the percentage of Np-237 in the target from 20% to 30% will be completed. There are approximately 40 positions available which would accommodate 80 targets. This could produce approximately 300 g of heat source material depending on position availability, target availability, and position qualification.

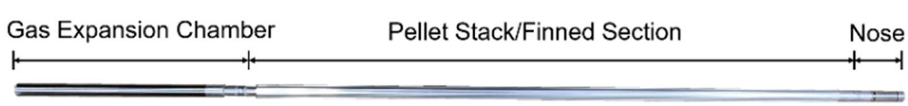

Figure 1. **An ATR GEN I target.**

II.F Upcoming cycle 173B irradiation

Cycle 173B is a 60-day cycle estimated to start in summer 2024. There are approximately 17 inner core positions available which would accommodate 34 targets and could produce approximately 120 g of heat source material. Analysis for the I positions will be in progress during this cycle. The analysis may include all of the medium and large I positions, depending upon the difficulty of completing a bounding analysis. There are four large I positions available that could include approximately 23 irradiation locations each, and nine medium I positions that would include seven irradiation locations each. This would allow for a maximum of 155 positions, accommodating 310 targets. Ultimately, this could produce 700 g of heat source material. Also note that these positions have an assay of approximately 94%, which is higher than the inner core positions. Producing this amount of material would require approximately six cycles which span approximately 2 years.

III. Qualification of ATR GEN I target in NEFT, SFT, Inner-A, and H positions

III.A Mechanical design

The basket design for the NEFT, Inner-A, and H positions utilizes features from existing basket designs. The main basket body is made from extruding, thin-walled, aluminum tube to create ridge features along the longitudinal axis that help keep the basket vertically centered within the flux trap. The head of each basket is designed to be used with hand tools to remove and manipulate each basket. The nose of the basket has been redesigned to allow for a stronger fillet weld, while still allowing for the optimal flow through the basket. Each basket allows for two targets to be stacked 'nose to nose.' This allows for 46 targets to be irradiated in the NEFT, fourteen in the SFT, and two targets per inner-A and H position.

III.B Neutronics analysis

The primary neutronics code used in qualifying the PFS ATR GEN I target design for the Inner-A and H positions was MC21 (Ref. 2). MC21 and its associated API, PUMA, are part of the common Monte Carlo design tool, CMCDT, provided for use in the ATR. MC21 was used to calculate neutron and photon heat generation rates during irradiation, fission gas production, Pu-238 production, fission density, and experiment reactivity. Due to the lack of development of the ATR model in MC21 for decay heat and dose consequence at the time calculations were performed, MCNP5 (Ref. 3 and 4) coupled with ORIGEN2 (Ref. 5) (MOPY) was used to calculate the decay heat and the dose consequence instead. MOPY (Ref. 6) works by using reaction rates and fluxes calculated by MCNP in an ORIGEN2 depletion on the pertinent materials. MC21 used ENDF-VIII.0 (Ref. 7) cross sections for all pertinent materials, while MCNP5 primarily used ENDF-VII.0 cross sections, with one exception being the use of TENDL-2017 (Ref. 8) for the Np-236m isotope.

The MC21 model of the neptunium target included 52 axial and five radial discretization's per target. An abbreviated PUMA model of a PFS target is shown in Figure 2. The PFS GEN I stack up includes two targets nose to nose at 5.25 in. and 0.125 in. above core centerline for the Inner-A and H positions, respectively. Existing hardware in the inner-A positions is what causes the targets in the Inner-A positions to be raised 5.25 in. above core centerline. The MCNP model utilized 40 axial and one radial section per target.

Neutronics analysis for qualification of the PFS ATR GEN I target in the Inner-A and H positions of the ATR was completed for a maximum central lobe power of 25 MW for a 65-day irradiation period. The nominal cycle length that the ATR targets will be irradiated for is 60 days. Qualifying the targets to a total of 65 days and to a maximum power of 25 MW allows for the targets to be qualified for multiple cycle lengths and powers under one analysis (Ref. 9). At the nominal 60 effective full power days (EFPD), the peak production positions are in the lower targets of A6 and H12.

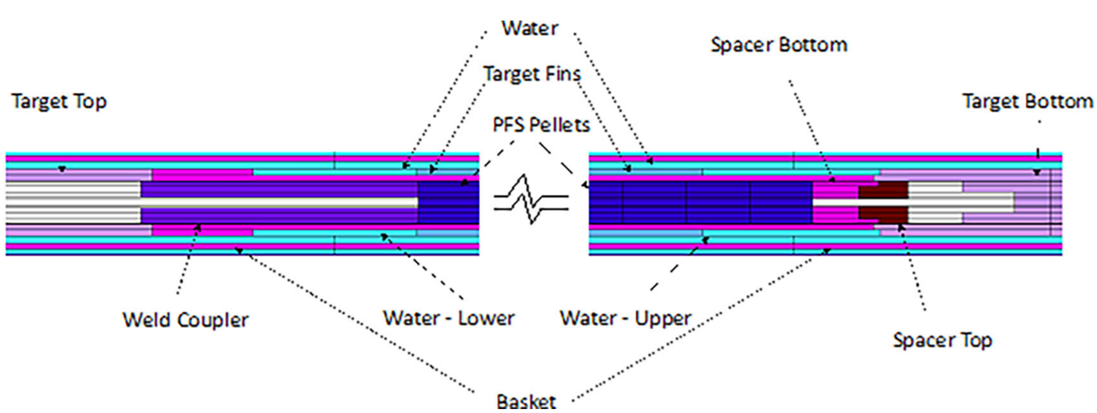

Figure 2. An abbreviated PUMA model of a PFS target.

Pu-238 production rate estimates for the peak positions were calculated to be 2.43E-03 g/MWd and 1.64E-03 g/MWd for the lower and upper target in A6, respectively; and 2.26E-03 g/MWd and 2.14E-03 g/MWd for the lower and upper target in H12, respectively. The inner-A and H positions were able to achieve a higher assay than the NEFT targets with the average assay being approximately 92% for the inner-A and H positions compared to the overall peak assay in the NEFT of approximately 88%. For reference, NASA RPS missions typically use heat source materials with a minimum assay of 82.5%.

Work is currently underway to qualify the SFT position for the ATR GEN I targets. The qualification is performed using the MCNP to ORIGEN2 in Python (MOPY) method. MOPY is a python-based tool that passes data between MCNP and ORIGEN2 for depletion and activation. The SFT GEN I analysis is being completed using ENDF-VIII.0 cross sections for all experimental materials.

The MCNP model used for the SFT qualification utilized 40 axial regions and one radial section for each neptunium target. Like the NEFT and inner-A and H positions, the SFT GEN I stack up will have two targets stacked nose to nose and elevated slightly above the core centerline to accommodate existing hardware.

Like the previous neutronic analysis, the SFT qualification assumes a 65 EFPD cycle as well as the projected peak power for the SFT of ATR. Preliminary analysis of the SFT is anticipated to produce 56 g of Pu-238, with an average peak Pu-238 Assay of 87.5% after 60 EFPDs of irradiation.

Qualification is also being done simultaneously on the EFT position of ATR. The SFT analysis will envelop the results for the EFT as the EFT and SFT have identical geometric configurations and the South Quadrant of ATR is typically operated at a higher power than the East Quadrant, which results in a higher flux and higher heating rate in the SFT as compared to the EFT.

Additional neutronic analysis was completed to support continued irradiation of the ATR GEN I targets in the NEFT. Due to an increase in the projected ATR power splits a full neutronic analysis was completed for the NEFT. This analysis demonstrated compliance to the safety requirements for continued irradiation. To support complete utilization of all Pu-238 targets located at INL, an analysis was also completed to qualify the irradiation of a single HFIR GEN II target in the SFT. The SFT was selected as it had previously been qualified for the irradiation of seven HFIR GEN II targets, and thus much of the original analysis is bounding to the single target.

III.C Thermal analysis

Thermal-hydraulic analyses were performed to support irradiation in the inner-A and H positions, as well as the NEFT. The qualification of the ATR GEN 1 targets in the NEFT included a parametric analysis of requisite safety scenarios under a wide range of thermal-hydraulic conditions. The results of the analysis provided system response surfaces for critical safety quantities such as DNBR, FIR, and peak component temperature (Figure 3).

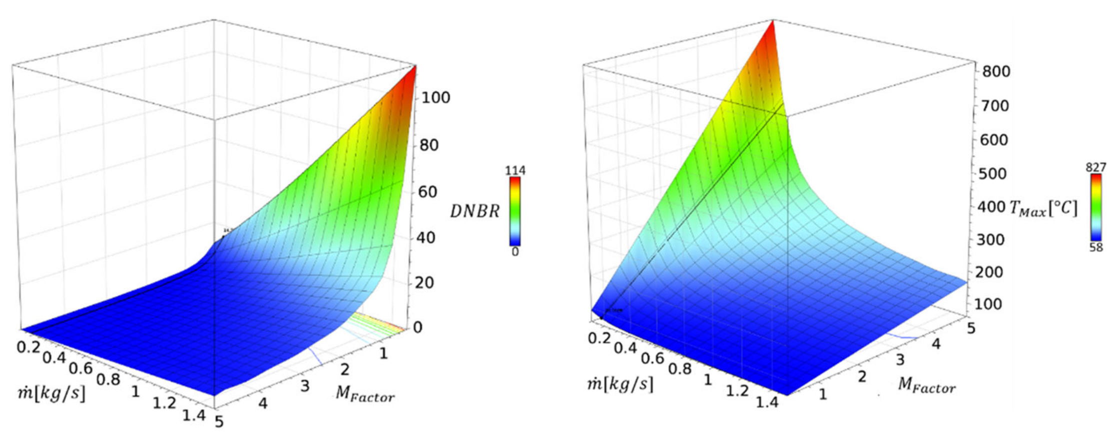

Figure 3. Response surfaces for the (a) Minimum DNBR and (b) maximum component temperature of the ATR GEN 1 target under various thermal-hydraulic conditions.

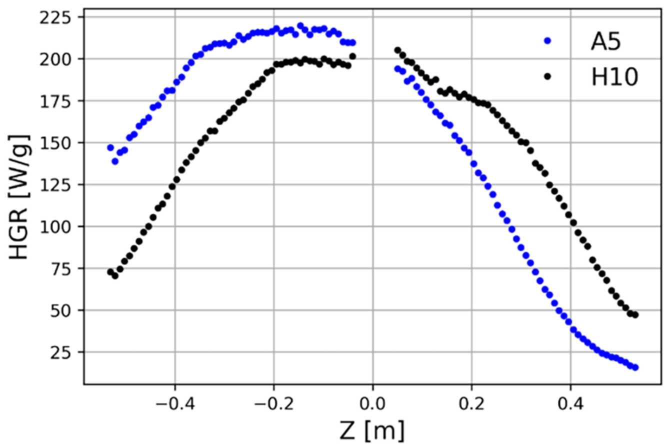

Figure 4. Axial heat generation rates of the NpO_2-Al cermet pellets at the end of a 65-day cycle for the A5 and H10 positions.

The response surfaces were used to facilitate the qualification of the Inner-A and H positions, by verifying that the mass flow rate and total heat rate for the most limiting positions were within the analyzed bounds. The heat generation rates for the pellet stacks in the most thermally limiting positions (A5 and H10 positions) are shown in Figure 4.

Flow rates for all analyses were provided using RELAP5-3D (v.4.4.2) (Ref. 10) while the subsequent thermal analyses were performed using ABAQUS (v.2018hf3) (Ref. 11) and STAR-CCM+ (v.16.06.010-R8) (Ref. 12).

While the NEFT featured similar analyses as the other irradiation locations, to support discharge of the experiment from the NEFT, a CFD analysis was performed to determine the number and configuration of targets to be removed in order to avoid disruption to the operational cadence. The analytical approach used features a simplified geometry, specifically a 2-D symmetric cross section at the axial center, as shown in Figure 5.

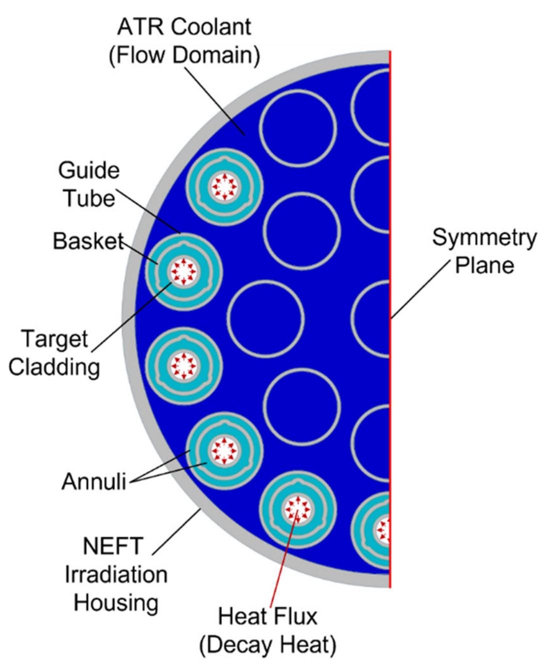

Figure 5. Schematic of the CFD model used for horizontal analyses of the PFS experiment in the NEFT housing.

The coolant was modeled as a solid between the cladding and baskets, as well as in between the baskets and guide tubes, thereby reducing analytical complexity exclusively to conduction in these regions. An effective thermal conductivity, accounting for heat transfer enhancement via free convection in these regions, was implemented, as shown in Eqns. 1 through 3. These were implemented via look-up tables in STAR-CCM+.

$$\frac{k_{Eff}}{k} = 0.386\left(\frac{Pr}{0.861+Pr}\right)^{1/4} Ra_c^{1/4} \tag{1}$$

$$L_C = \frac{2\left[ln\left({}^{r_o}/_{r_i}\right)\right]^{3/4}}{\left(r_i^{-3/5}+r_o^{-3/5}\right)^{5/3}} \tag{2}$$

$$Ra_C = \frac{g\beta(T_w - T_\infty)L_C^3\rho^2 C_p}{k\mu} \tag{3}$$

Equation 4 presents the Nusselt number correlation used to estimate the heat transfer from the guide tube surface to the flow domain.

$$Nu_{Ra} = \left[0.6+\frac{0.387Ra^{1/6}}{\left[1+\left({}^{0.559}/_{Pr}\right)^{9/16}\right]^{8/27}}\right]^2 \tag{4}$$

The results of this analysis predict a maximal coolant temperature of 85°C (185°F), with the general trend that the maximal coolant temperatures are inversely related to the gravitational field; this aligns with expectation.

A supporting calculation was performed using RELAP5-3D. Certain conservatisms were not implemented in the RELAP5-3D simulation, as exploratory calculations indicated a mixed convective-conductive heat transfer modality was observed between the basket and guide tube. Figure 6 presents the nodalization implemented in RELAP5-3D.

Calculating a decay heat curve appropriate for the time scale and implementing the Nusselt number correlation presented in Equation 4 via 'htc-temp' tables in RELAP5-3D, resulted in the temperature traces shown in Figure 7. The temperature curves depict a maximum temperature within the region between the target and basket of 76°C (168°F), well within expectation, given the increased heat transfer implemented in the RELAP5-3D model.

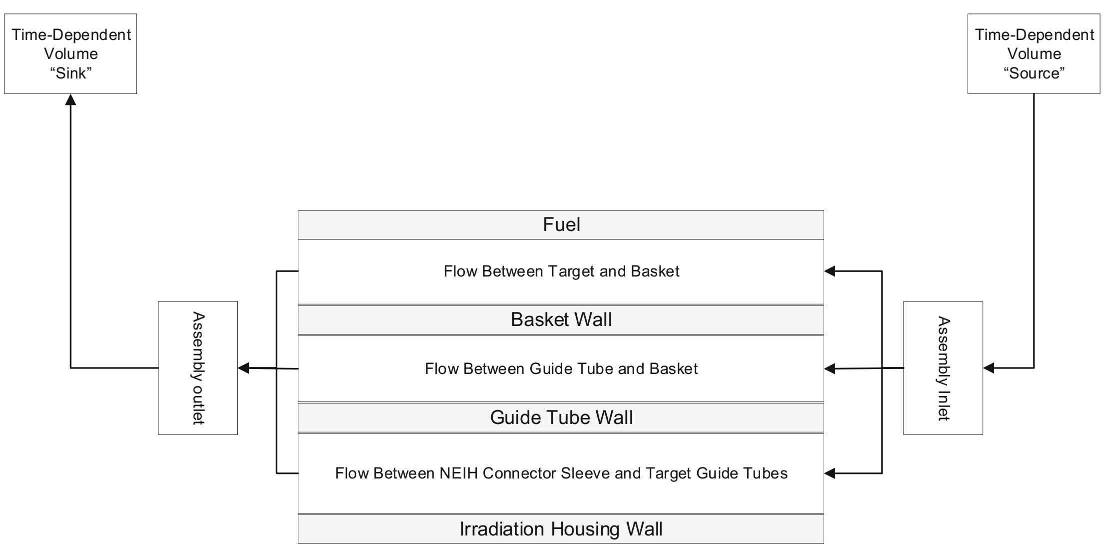

Figure 6. Schematic showing RELAP5-3D component and heat structure orientation.

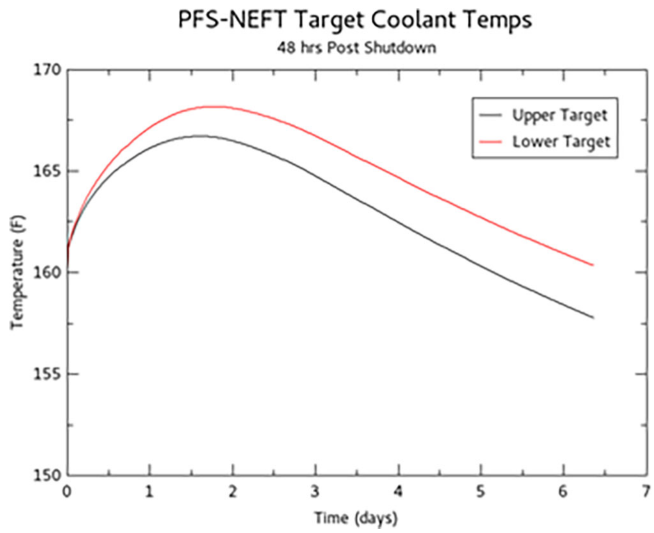

Figure 7. **Upper and lower target coolant temperatures calculated in RELAP5-3D.**

III.D Structural analysis

Structural analyses were performed to support irradiation in the NEFT, the inner A and H positions. Limits for temperature and pressure (both internal and external) for the target were determined using the acceptance criteria defined in the American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel (B&PV) Code. Based on the low internal pressure of the target, the requirements of ASME Section III, Class 3 vessels were used as a guide. The qualification of these targets in the three positions included a comparison of these limits to those calculated in the thermal analysis. Other loadings, such as handling loads from transferring components to and from the reactor, were also considered. These include an accidental drop of the target through water from a height of 45 ft which could occur at the deepest portion of the ATR canal.

III.F ATR safety considerations

Along with existing Experiment Safety Analysis (ESA) already developed for both the PFS experiment in the I-7 and SFT positions, an additional ESA was developed for the ATR GEN I target irradiations in the NEFT, inner-A, and H positions. This ESA utilizes the analyses performed, as previously discussed (neutronics, thermal, and structural), to demonstrate that the new PFS GEN I targets can be irradiated in the ATR in compliance with the technical safety requirements and the approved authorization safety basis, established by ATR's SAR. The GEN I ESA was also developed and authorized under an ATR Complex procedure that addresses experiment receipt, handling, reactor loading, irradiation, discharge, storage, preparing for shipping from ATR, and waste disposal. The PFS ATR GEN I ESA demonstrates that operation of the PFS experiments are in accordance with the restrictions identified in the ESA and within the authorization safety basis of the ATR.

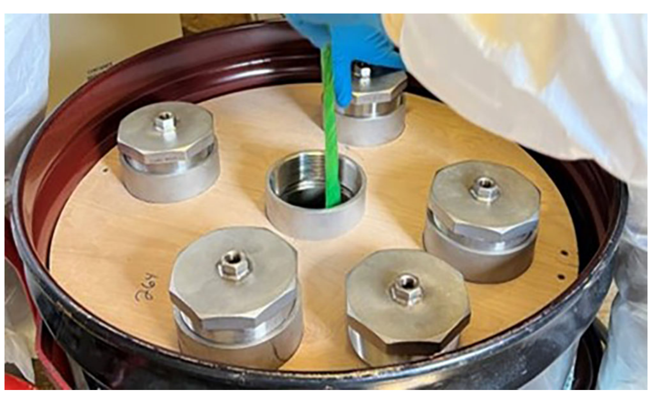

Figure 8. **An ATR GEN I target being loaded into the shipping container.**

IV. Increasing NP concentration in targets

IV.A Preliminary neutronic analysis

Work has begun on utilizing MC21 to analyze a potential increase in the Np-237 concentration from 20 vol% to 30 vol%. Preliminary findings indicate increasing the Np concentration to 30 vol% would result in a potential Pu-238 production increase in the range of 20-30%.

V. Shipments

INL has received four shipments of targets from ORNL, consisting of 85 gal drums. Originally, shipments were made by inserting one target per drum. This allowed for a maximum of eight targets to be shipped to INL at a time. An updated design to the drums has allowed five targets to be shipped per drum with a maximum of 40 targets per shipment. The updated design was completed to support the need to ship and receive approximately 200 targets per year. This design uses five 4 in. stainless steel pipes supported in the 85 gal drums, as shown in Figure 8.

VI. Conclusions

INL has successfully completed qualification for the ATR GEN I targets in the NEFT, inner A, and H positions. This included using a CFD and RELAP5-3D analysis to support unloading the NEFT within operational time limits. Efforts on requalifying the SFT and qualifying the EFT for the ATR GEN I target are in the final stages of completion. With the qualification of more positions and increased efficiency in shipments, INL is on track to meet production goals by 2026.

Data availability

Due to the sensitivity of production of plutonium in nuclear reactors and the use of Pu-238 in space power sources, some codes, source files and intermediate data sources have been identified as having distribution restrictions due to International Traffic in Arms Regulations (ITAR) and Export Controlled guidance.

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Open Peer Review

Current Peer Review Status: ✔

Version 1

Reviewer Report 11 October 2024

https://doi.org/10.21956/nuclscitechnolopenres.18778.r27670

✔ **Qingquan Pan**
Shanghai Jiao Tong University, Shanghai, China

The reviewer also works on the production of plutonium-238 (Nuclear Science and Techniques, 2024, 35, 88.), but only at the level of theoretical calculations. I am very envious of the authors for having a research reactor, which allows them to conduct related experimental studies.
This paper is a high-quality research paper that provides important technical and management insights into the production of Pu-238. I support the indexing of this paper and look forward to the authors' future progress.

The review recommendations are as follows:
(1) The overall quality of the paper is high and no major revisions are required. The authors are advised to carefully proofread the text and figures in the final version to ensure accuracy.
(2) Further details and data on the experiments could be added to enhance the reproducibility and validity of the study.
(3) Given the sensitivity and safety of Pu-238 production, the authors are advised to strengthen the assessment and discussion of safety risks in the discussion section to ensure the compliance and safety of the study.

Is the background of the case's history and progression described in sufficient detail?
Yes

Is the work clearly and accurately presented and does it cite the current literature?
Yes

If applicable, is the statistical analysis and its interpretation appropriate?
Yes

Are all the source data underlying the results available to ensure full reproducibility?
Yes

Are the conclusions drawn adequately supported by the results?

Yes

Is the case presented with sufficient detail to be useful for teaching or other practitioners?
Yes

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: reactor physics

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.