INL/CON-23-75787-Revision-0

Pu-238 Production Progress at Idaho National Laboratory From December 2022 to December 2023

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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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Idaho National Laboratory (INL) has an ongoing effort to produce Pu-238 for NASA deep space missions. Recent work at INL has consisted of irradiation of Pu-238 production targets in the Advanced Test Reactor (ATR), generating more than an estimated 400 grams of Pu-238 heat source material between March and October 2023. Additionally, INL began qualifying Pu-238 production targets with a higher loading of Np-237 to further increase the production of Pu-238 production in later years.

INL has also updated the analysis as a result of operational changes at ATR. One instance was updating the analysis to enable a single Pu-238 production target to be run in ATR's South Flux Trap (SFT), rather than the previous seven production targets, to make use of a spare target from a discontinued design. Higher lobe powers in ATR were also analyzed due to potential changes in planned lobe powers.

I. Introduction

The plutonium production effort at INL has been tasked with contributing to the 1.5 kg per year constant rate production goal of Pu-238 heat source material in the United States by 2026. The Pu-238 heat source material is an oxide form to be used as fuel in Radioisotope Power Systems that enable deep space NASA missions. To achieve the production goal, the program has focused on qualifying as many positions in the inner core of the Advanced Test Reactor (ATR) as possible (see Fig. 1). This has allowed for Pu-238 production to become a backup for other experiments and fill in gaps in ATR's core loading plans. The other benefit is the more positions that the targets are inserted into the reactor for each cycle, the greater the Pu-238 production.

This paper will focus on the progress made during the last year. Work began with completing the qualification on inserting the ATR GEN I Target with 20% Neptunium (Np) into the south flux trap (SFT). This is the final inner core position left to qualify for 20% Np targets. Work also continued on completing the qualification for 30% Np.

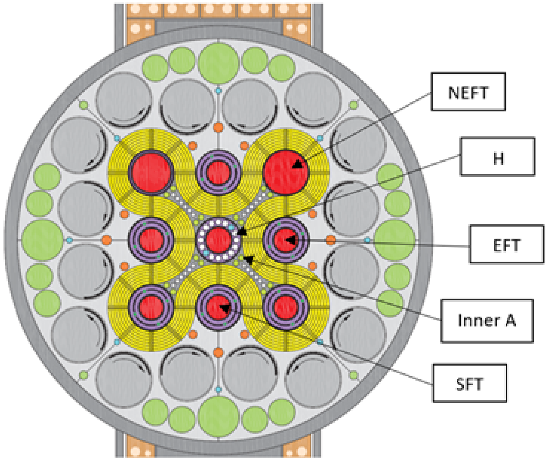

Fig. 1. Cross-section of ATR with target positions identified.

II. Pu-238 Production at INL

II.A. Cycles Completed in 2023

Two irradiation cycles were completed in 2023. The first cycle was 171A (54 days), where a total of 57 targets were irradiated in the positions shown in Table 1. The second cycle was 171B (64 days), where a total of 49 targets were irradiated in the positions shown in Table 1.

TABLE 1. Targets Irradiated in ATR during CY 2023

ATR Core Position	Cycle 171A	Cycle 171B
NEFT	46 ATR GEN I	46 ATR GEN I
SFT	7 HFIR GEN II	1 HFIR GEN II
Inner A	2 ATR GEN I	0 Targets
H	2 ATR GEN I	2 ATR GEN I
	Total	**106**

Between these cycles, 106 targets were successfully irradiated in ATR which generated approximately 450 grams of heat source material. These targets are currently being stored in ATR's canal awaiting shipment.

II.B. Upcoming Cycles

For the upcoming calendar year, 2024, there will be three 60-day cycles. The cycles are listed in Table 2. For cycle 173A and 173B, targets will only be inserted in the North East Flux Trap (NEFT).

TABLE 2. Planned Target Irradiation in ATR during CY 2024

ATR Core Position	Cycle 173A	Cycle 173B	Cycle 175A
NEFT	46	46	0
Inner A	0	0	10
H	0	0	16
		Total	**118**

With the upcoming cycles, it is expected that a total of 118 targets will be irradiated. It is estimated that these targets will make about 500 grams of Pu-238 heat source material. Irradiated targets will need to stay in ATR's canal for a minimum of six months.

III. Qualifications Efforts

III.A. SFT 20% Np Target Qualification

To satisfy the safety requirements for irradiating the ATR GEN I targets in the SFT of ATR, multiple neutronic calculations were completed. The SFT is one of the most challenging positions in ATR to qualify due to typical power levels creating challenging thermal and neutronic conditions. Nominal values of ATR operating conditions were used to model the ATR GEN I targets in MCNP, and the results were scaled to the projected peak power splits. To properly capture the axially dependent behavior of the neptunium pellet material in each target, each pellet stack was divided into forty axial segments.

MCNP5, a general-purpose Monte Carlo N-Particle transport code, was used to calculate the pertinent neutron and photon heat generation rates within all experiment materials. MCNP was also used to calculate the neutron fluxes and reaction rates for pertinent reactions on the neptunium pellet material. This information was then passed into ORIGEN2 to deplete the neptunium pellet material. The ENDF/B-VIII.0 cross section library that comes with MCNP was used to perform the MCNP calculations. The standard ATR cross section library was used for ORIGEN2 along with MCNP-calculated replacement cross sections. A python-based code, MCNP to ORIGEN2 in Python (MOPY), was used to extract the fluxes and reaction rates calculated from MCNP.

In addition to the calculations listed above, and to further demonstrate reactivity safety compliance, the reactivity worth of the unirradiated experiment and the end of cycle experiment were calculated using the 19-plate MCNP model of ATR.

After 60 days of irradiation, the Pu-238 average assay was calculated for each target located in the SFT. The peak average Pu-238 assay is 87.49%. Using MOPY it is estimated that approximately 56 grams of Pu-238 will be produced in the SFT of ATR during a 60-day cycle.

The thermal qualification of the 20% Np targets in the SFT/East Flux Trap (EFT) was sufficiently bounded by the analysis performed for the 30-35% Np loading in the same position. The total heat rate, peak heat rate, and decay heat rates were all below the 30-35% cases at the same hydraulic conditions. Therefore, the results of the 30-35% qualification, as well as the experiment handling times, were directly applicable to the 20% Np loading.

The purpose of the structural safety analysis was to evaluate the target and its associated hardware under various potential structural loading scenarios to ensure the safety of operational personnel and the public. The structural loadings considered in this evaluation, while within the ATR, included the following: internal pressure within the target due to the release of fission gas, external pressure, external pressure differential (acting on the length of the assembly), pressure and skin friction drag forces due to coolant flow velocities, flow induced vibrations, thermal loads, and cyclical loads. The decision for which structural loading scenarios were to be evaluated in the structural analysis was based upon the probability of the event occurring and the desired state of the structural components after each event. These events included normal reactor operation, a flow coast down event due to loss of commercial power, a reactivity insertion accident for in-pile tube voiding, overpressure, and a loss of coolant accident. Events with low probability of occurrence, and when the consequence of a pressure boundary losing its integrity meets the safety limits defined by INL's safety analysis report (SAR), were excluded from the structural evaluation. Other structural 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.

For the structural analysis to be useful for multiple positions within the ATR, limits for temperature (peak and gradient), pressure (internal and external), and coolant flow velocities were established. The response of each structural component (i.e., stress, strain, deformation, etc.) under these limiting conditions was calculated using, where simplifications could be made, hand calculations or, where simplifications could not be made, using the finite element software ABAQUS. These responses were compared to acceptance criteria. For the non-pressure retaining components, this criterion was typically the yield strength of the material at temperature. Due to the potential of fission gas release, the target was treated as a pressure vessel. Acceptance criteria limits defined in the American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel (B&PV) Code were used. Though other acceptance criteria could be used, this code was used because it provides a nationally accepted design/analysis approach which INL has used and adapted to various nuclear experiments. Based on the low internal pressure of the target, the requirements of ASME Section III, Class 3 vessels were used as a guide. The limits of temperature, pressure, and coolant velocities, using these acceptance criteria, were compared to those calculated in the thermal analysis and from the design specification of the ATR. For the SFT, these values were within these calculated limits and each structural component was considered to meet the safety requirements and allowed into the ATR.

III.B. 30% Np Target Qualification in the NEFT, SFT, EFT, Inner A and H

Analyses were done to qualify an ATR GEN I target with both a 30 vol-% Np target and a 35 vol-% Np target using the common Monte Carlo design tool (CMCDT). CMCDT consists of the Physics Unified Modeling and Analysis (PUMA) API, version 9.1.1, and the Monte Carlo code, MC21, version 9.00.02. ORIGEN, as part of the SCALE6 package, was used for the decay heating and source term calculations. Nominally, the desired target concentration will contain 30 vol-% Np with analyses also completed at 35% Np to account for variances in the pellet manufacturing process. The analyses could not be completed at 35% Np alone due to nonlinearity in the safety case between 30% and 35%. The entire inner core, consisting of the NEFT, SFT, EFT, inner-A and H positions, were qualified at both Np concentrations. Each position contains two targets stacked nose-to-nose, and there are 23 positions in the NEFT, seven positions in the SFT, seven positions in the EFT, eight individual A positions, and 14 individual H positions, all of which required analyses at both the 30 vol-% and the 35 vol-%. Overall, 164 targets were analyzed to meet the qualification. Fig. 2 shows the modeled stackup.

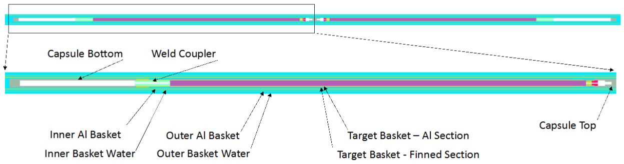

Fig. 2. ATR GEN I Target Stackup

Due to self-shielding in the targets, the overall wattage produced is largely the same regardless of Np loading, with the notable exception being at the center where the two ATR GEN I targets meet. This center has a high Np loading resulting in more peaking along the centerline. Fig. 3 shows the calculated watts for the peak position of the SFT irradiated at 25.7 MW. It can be seen that the centerline peaking and lower edge effects are highest for the 35% Np, followed by 30% Np, and the lowest peaking for the 20% Np. However, the overall wattage is largely similar, but the highest across the curve is typically the 20% Np with the lowest over the curve being the 35% Np.

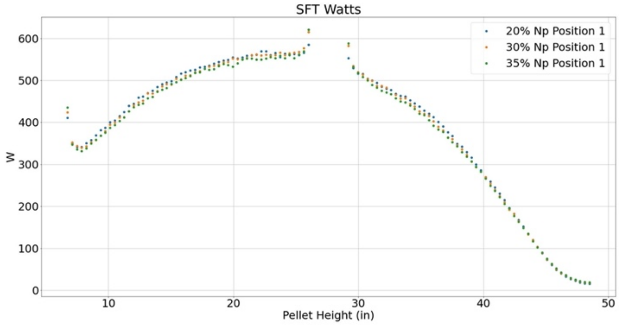

Fig. 3. Heating comparison (W) for 20%, 30%, and 35% Np concentrations in the SFT position 1 shows similar heating regardless of Np concentration.

Because an increase in Np results in a decrease of aluminum, pellet mass increases as the amount of Np increases, resulting in heat generation rates (W/g) decreasing as the Np concentration increases. Table 3 gives averages for production rates and Np conversion parameters for each of the individual positions. Each position is scaled to its respective bounding lobe power – 22 MW in the NE, 25 MW for the inner-A and H positions, 25.7 MW in the S, and 23.7 in the E. Over a full 60-day cycle, the NEFT has a maximum total production of 202 g Pu-238, the SFT has a maximum total production of 71.6 g Pu-238, and the EFT has a maximum total production of 67.4 g Pu-238.

Position	Avg. Pu-238 (g)	Avg. Assay (%)	Avg. Pu-236 content (ppm)	Lobe Power (MW)	Number of Positions
EFT	4.82	89.5	4.54	23.7	7
SFT	5.11	88.9	4.67	25.7	7
NEFT	4.39	90.9	5.60	22	23
H	4.15	93.3	6.77	25	14
Inner-A	3.90	93.7	8.04	25	8

TABLE 3. Position Average Production Values at 60-days for a Target with 30 vol-% Np.

The thermal qualification of the 30-35% Np loaded ATR GEN I targets was accomplished by leveraging previous analyses and utilizing a combination of finite element analysis (FEA) using ANSYS and ABAQUS and thermal/hydraulic system codes (RELAP5). The general safety analysis for the ATR GEN I target performed in the qualification of the 20% Np targets was used to facilitate the qualification of the 30-35% Np loaded targets. The heat rates for the A and H positions were well within the applicable bounds of the generalized analysis. However, the high heating rates experienced in the SFT and the reduced flow rates in the NEFT required further analysis to demonstrate compliance with requisite safety requirements for irradiation in ATR. Higher fidelity FEA models were developed using ANSYS (v.2022R2) based on the targets with the highest heat generation rates in the SFT (Target 1) and NEFT (Target 23) provided by neutronics analyses described above. The ANSYS models utilized thermal fluid elements to include the local increase in bulk fluid temperature along the length of the target. The mass flow rate for each thermal fluid element was prescribed based on the results from the RELAP5 model. The results provided sufficient margin for peak temperature, internal pressure (including fission gas release), minimum departure from nucleate boiling ratio (DNBR) and flow instability ratio (FIR) in the case of a flow coast down (FCD) and condition 2 reactivity insertion accident (RIA-2).

Post irradiation, the reactor is depressurized and flow is reduced and eventually shut off to allow for removal of the experiments from the ATR. The timeline for depressurization and shutdown is largely determined by the decay heat of the experiments within the reactor. A RELAP5 model was used to determine how long forced flow cooling must be maintained until the targets may be passively cooled by natural convection. The NEFT required the longest cooling time due to the number of targets followed by the SFT/EFT and A and H positions, respectively.

The required cooling time for the experiment via natural convection in air and water must be assessed for handling times or in the case that the experiment is dropped to the canal floor and/or a canal draining event. These analyses are used in the experiment safety analysis report to inform operations at ATR on safe handling times for the experiment. Following reactor shutdown and the requisite forced flow cooling time, the experiment remains in the reactor until it is safe for handling.

In the horizontal-in-air and water scenarios, the experiment assembly was modeled within the basket and immersed in the desired medium (water or air) using a 1-D thermal resistance network. A schematic of the resistance network, overlayed on the ATR GEN I geometry, is shown in Fig. 4.

The linear heat rate was calculated from the total decay rate of the hottest target stack of the NEFT, SFT, and A and H position with 35% Np pellets. The coolant between cladding-basket was modeled as a solid, simplifying this portion of the analysis to conduction. The thermal conductivity for these regions was replaced by an effective thermal conductivity to account for the heat transfer enhancement due to free-convection between two concentric cylinders. In all irradiation positions, the experiment was determined to be safe for removal from the reactor following 18 hours of decay time, and safe for transportation after six months in the ATR canal.

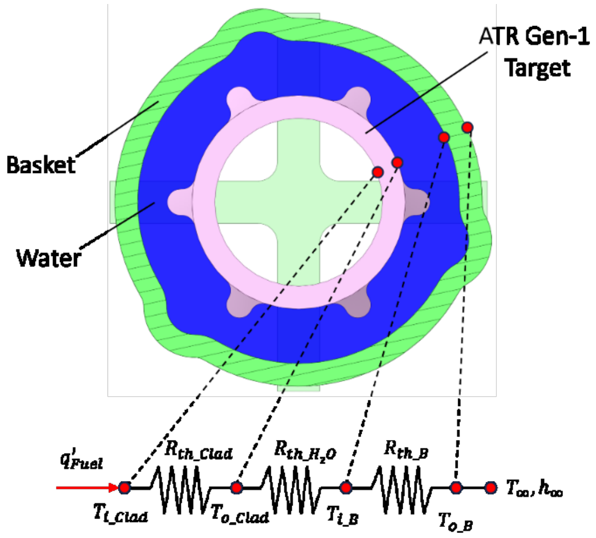

Fig. 4. Thermal resistance network of the ATR GEN I target for a horizontal in water analysis.

Due to the increase in thermal load of the 30% Np, there needed to be an increase in flow through the SFT housing. The orifice under each basket was increased from 0.25-in. to 0.5-in. diameter.

As was with the SFT 20% qualification, a structural safety analysis was performed for the 30% Np targets for the various positions (NEFT, SFT, EFT, Inner A and H). The previously determined limits on temperature, pressure, and coolant velocities were compared to those calculated in the thermal analysis using the 30% Np targets. These values were within the calculated limits and each structural component was considered to meet the safety requirements and allowed into the ATR.

III.C. Single SFT Target and Power Increase in the NEFT

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. Similar to the 20% GEN I SFT neutronic analysis, MCNP5, a general-purpose Monte Carlo N-Particle transport code, was used to calculate the pertinent neutron and photon heat generation rates within all experiment materials. MCNP was also used to calculate the neutron fluxes and reaction rates for pertinent reactions on the neptunium pellet material and this information was passed into ORIGEN2, via MOPY, to deplete the neptunium pellet material.

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 in ATR.

The resulting heat generation rates for a single HFIR GEN II target in the SFT were compared against a previously successful thermal qualification of a fully loaded SFT configuration (seven total assemblies). The overall thermal load from the single target was sufficiently bounded by the previous analysis and, therefore, met all requisite safety requirements for irradiation in the ATR.

The limits on temperature, pressure, and coolant velocities, previously determined, were compared to those calculated in the thermal analysis using a single target. These values were within the calculated limits and each structural component was considered to meet the safety requirements and allowed into the ATR.

IV. Shipping and Storage Updates

IV.A. Target Receipts

Fiscal year 2023 saw two separate deliveries from ORNL of Neptunium targets to advance the Pu-238 production efforts. These deliveries contained 25 targets in February and an additional 24 in March. These targets were designed for the ATR, thus requiring two targets to be stacked end to end to better utilize the ATR at INL. There are several locations where these targets may be stored at INL, but the intention is to place them into ATR as soon as possible. At this point there are very few positions in the ATR that are not qualified for this purpose. Once the targets are received and stored, it is expected that the shipping containers be reloaded and returned to ORNL on the same delivery transportation as they were received.

IV.B. Gamma Activity with Increased Decay Time

The isotopic gamma activity for the irradiated NpO_2-Al material composition was investigated for increased decay time, following discharge from the ATR. The NpO_2-Al material composition was assumed to be irradiated for 65 days in the ATR. Also, the NpO_2-Al material composition was assumed to contain 20 vol-% NpO_2.

The base case for this study assumed 180 days decay time, the same decay time that was assumed for the original SAR prepared by ORANO. Next, the decay time was increased to 365 days. The increase in decay time produced a significant reduction (greater than 40%) in isotopic gamma activity. The reduction in isotopic gamma activity is primarily associated with fission products which have half-lives less than 1 year. Therefore, the decrease in the isotopic gamma activity will also occur for higher volume fractions of NpO_2.

It should be noted that the total dose from the Battelle Energy Alliance Research Reactor (BRR) transportation package is now dominated by the contribution from neutrons. The updated SAR will reflect the contributions provided by neutrons and gammas.

IV.C. Calculated Maximum Number of Targets for SAR Update

Significant improvements have been made to increase Pu-238 production from the Pu-238 producing targets irradiated in the ATR. First, the target design was changed from the HFR GEN II to the ATR GEN I target design. This change allows two ATR GEN I targets to be irradiated in a single ATR experiment location. This change significantly increases the annual Pu-238 production from the ATR.

Second, the volume fraction of NpO_2 in the NpO_2-Al material composition was increased from 20 to 30 percent. Increasing the initial Neptunium content in the target leads to an increased Pu-238 content from the target at discharge from the ATR. Third, the ATR GEN I targets were irradiated in ATR core locations with higher powers than those assumed in the original analyses for the HFIR GEN II target design.

The original dose analysis performed for the BRR transportation package assumed 96 HFIR GEN II targets. These targets were assumed to be irradiated in the Inner A positions of the ATR.

The NpO_2-Al material composition in the HFIR GEN II targets were assumed to be irradiated for 65 days in the Inner A positions of the ATR. The isotopic composition for the NpO_2-Al material composition was then allowed to decay for 180 days after discharge from the ATR. The neutron and gamma source terms in the original SAR were then calculated based upon these assumptions.

Revised neutron and gamma source terms have been calculated based upon the discussed changes to the target design and irradiation positions. The updated SAR will contain the limiting neutron and gamma source terms for NpO_2 volume fractions from 20 to 30 percent and for targets irradiated in any ATR core location. The maximum number of ATR GEN I targets which can be safely shipped in the BRR transportation package will be presented in the updated SAR.

V. ATR Operations Updates

V.A. Targets Stored in ATR Canal

ATR started irradiation of the HFIR GEN II targets during the 166A-1 cycle with seven targets in the I-7 position. Since that time, ATR has irradiated HFIR GEN II targets in the SFT during the 169A, 171A, and 171B cycles. ATR has also irradiated ATR GEN I targets in the NEFT, H, and inner A positions during the 171A-1 and 171B-1 cycles. ATR recently complete the 171B-1 cycle, but since the first targets were irradiated in the 166A-1 cycle, ATR has irradiated 120 Pu-238 producing targets. These targets are currently being stored in the ATR canal until they can be shipped to ORNL for processing.

V.B. Safety Analysis Updates

The process of showing compliance to safety requirements for handling, storage, irradiating, and preparing targets for shipping experienced several process improvements during the past calendar year. The most prominent improvements dealt with preparing targets to ship from ATR after the desired irradiation time was achieved. A cask list for ATR was completed, which currently includes the safety requirements for shipping in the BRR (and GE-100) casks or transportation packages and acts as the top level of the shipping safety documents. Each transportation package has general requirements derived from the ATR experiment safety analyses (ESAs), or non-safety analyses. The purpose of the cask list is to organize and implement these requirements for the different casks used at ATR for experiments.

The cask list hands off to a shipping compliance (cask-specific) which utilizes an experiment configuration checklist (cask-specific) to verify compliance to the safety requirements prior to the shipment leaving ATR. This process improvement has streamlined the preparation to ship at ATR by referencing the necessary calculations/evaluations (e.g., thermal, as-run data, source term, hazard categorization) to demonstrate and verify compliance with the required safety requirements supporting these shipments. Future revisions to the cask list will include adding any outlying safety basis (SAR/TSR) requirements.

VI. Conclusion

During the past year, the team has made great advancements in the progress of contributing to the overall program goal. The team was able to qualify the final possible inner core positions, the SFT and EFT, for 20% Np ATR GEN I targets. The team was also able to complete the qualification for 30% Np ATR GEN I targets for all the inner core positions, NEFT, SFT, EFT, Inner A and H positions.

ACKNOWLEDGMENTS

This work was funded by the NASA Interagency Agreement # NNH19OB05A and DOE contract DE-AC05-00OR22725 and DE-AC07-05ID14517.

This work 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.

REFERENCES

1. F. Brown, B. Kiedrowsky, et. al, “MCNP5-1.60 Release Notes,” LA-UR-10-06235 (2010).
2. X-5 Monte Carlo Team, “MCNP-A General Monte Carlo N-Particle Transport Code, Version 5,” Volume I, LA-UR-03-1987, Los Alamos National Laboratory, April 24, 2003 (Revised 10/3/05) and Volume II, LA-CP-03-0245, Los Alamos National Laboratory, April 24, 2003 (Revised 10/3/05) (Vol. II is available with a licensed copy of MCNP).
3. A. G. Croff, “ORIGEN 2: A Versatile Computer Code for Calculating the Nuclide Compositions and Characteristics of Nuclear Materials, Nuclear Technology,” Vol. 62, pp. 335-352, 1983.
4. M.B. Chadwich et al., October 2006. “ENDF/B-VII.0: Next Generation Evaluated Nuclear Data Library for Nuclear Science and Technology,” UCRL-JRNL-225066,
5. TENDL-2017, “A.J. Koning and D. Rochman, "Modern Nuclear Data Evaluation With The TALYS Code System,” Nuclear Data Sheets 113 (2012) 2841.
6. GDE-594, “Experiment Design and Analysis Guide – Neutronics & Physics,” Rev. 2, (Internal Report) January 11, 2017.
7. B. G. Schnitzler, BGS-6-91, “Origen2 Cross Section Library Assessment for ATR Applications,” (Internal Report), Idaho National Laboratory, April 1991.
8. “Experiment Analysis Documentation / MOPY,” (Internal Report), Idaho National Laboratory, January 2022.
9. D. A. Brown *et al.*, “ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data,” *Nucl. Data Sheets*, vol. 148, pp. 1–142, Feb. 2018, doi: 10.1016/j.nds.2018.02.001.
10. “SCALE Code System,” Oak Ridge National Laboratory, Oak Ridge, TN, Government Manual ORNL/TM-2005/39 Version 6.2.3, Mar. 2018. [Online]. Available: https://www.ornl.gov/sites/default/files/SCALE_6.2.3.pdf
11. David P. Griesheimer, et. al, “MC21 v.6.0 – A continuous-energy Monte Carlo particle transport code with integrated reactor feedback capabilities,” Annals of Nuclear Energy, vol. 82 pp. 29 – 40, 2015.