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Changing the World's Energy Future

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Recent Pu-238 Production Activities at Idaho National Laboratory

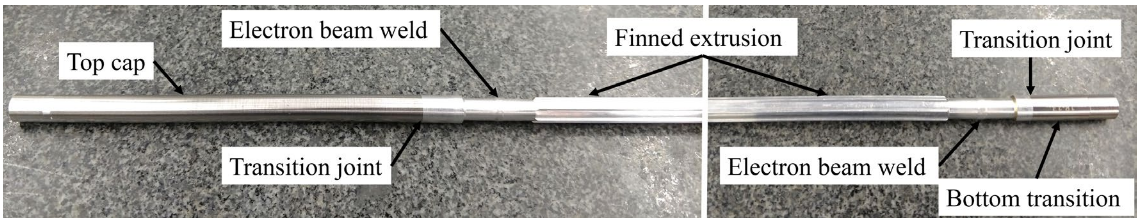
The Plutonium-238 (Pu-238) production program at Idaho National Laboratory (INL) is actively qualifying irradiation targets containing Neptunium-237 (Np-237) for the Advanced Test Reactor (ATR) to produce Pu-238 for future NASA missions. INL qualified and loaded seven targets in ATR's South Flux Trap (SFT) for cycle 169A, which occurred in Spring 2021. The irradiation qualification program has expanded to additional ATR irradiation positions after two baseline production targets in three positions validated significant production of Pu-238. The validation model was followed by the PFS-1 experimental test in the ATR Critical (ATR-C) Facility which verified Pu-238 production cross sections. This paper outlines the progress and status of the Pu-238 production program at INL. The qualification effort, safety analysis, hardware status, and future activities for qualification of an updated target design for use in the ATR will be discussed.

Keywords: space power, Pu-238, Advanced Test Reactor

I. PROGRAM OVERVIEW AND ATR IRRADIATION POSITION OVERVIEW

The Department of Energy's Constant Rate Production (CRP) program ensures a constant production of heat sources for future Radioisotope Power Systems (RPS). The goal of this program is to better support NASA nuclear enabled missions by providing stable staffing and production of materials for RPS. In the past, staffing was reduced and production was minimized between NASA missions. CRP reduces program risk by eliminating the significant challenges of having to hire and requalify staff, which can take years, having to maintain or restart equipment and infrastructure, and ensures an on-hand supply of heat sources, versus just in time support to NASA missions. One part of the CRP program ensures continued production of new Pu-238. New production relies on reactors at both Oak Ridge National Laboratory (ORNL) and Idaho National Laboratory (INL). Idaho National Laboratory (INL) is actively qualifying irradiation targets containing Neptunium-237 (Np-237) for the Advanced Test Reactor (ATR) to produce Pu-238 for future NASA missions. INL qualified and loaded seven targets in ATR's South Flux Trap (SFT) for cycle 169A, which occurred in Spring 2021. The irradiation qualification program has expanded to additional ATR irradiation positions after two baseline production targets in three positions validated significant production of Pu-238 [ref. 1]. The validation model was followed by the PFS-1 experimental test in the ATR Critical (ATR-C) Facility which verified Pu-238 production cross sections [ref. 2]. Continued supply of Pu-238 for RPS heat source production is an important part of ensuring the availability of future RPS as it sets the beginning of life thermal and electrical characteristics of the power supply. [ref 3]

The Np-237 target for Pu-238 production was designed to meet reactor insertion and nuclear quality assurance requirement for irradiation in ORNL's High Flux Isotope Reactor (HFIR) [ref. 4]. The target capsule is predominantly fabricated from Al-6061 extrusion and 304-L stainless steel. The target contains ceramic/metallic (cermet) feedstock pellets that consist of mixed and pressed aluminium and NpO_2 powders. The target capsules are sealed containing inert gas to enable sufficient heat transfer, eliminate deleterious chemical reactions within the target, and verify target hermeticity through non-destructive examination. Various design attributes, such as autogenous weld joints and bimetallic transition joints, are employed to improve fabrication reliability.



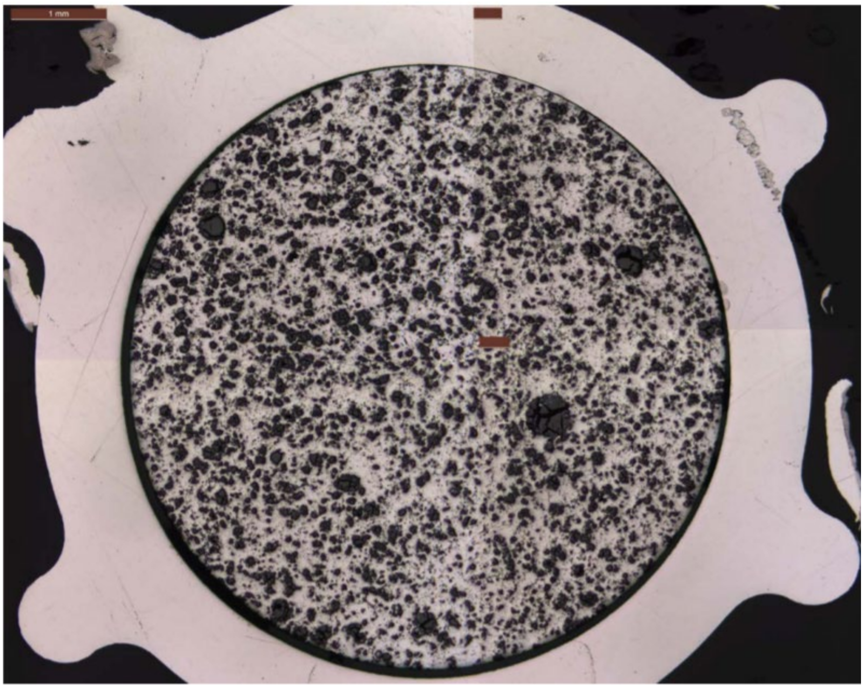


Fig. 1. Containment sub assembly for the production target, identifying specific design features(top) [ref. 4], and a cross section of the finned extrusion with a pellet (bottom) [ref. 5.].

The INL team has qualified the I-7 and SFT positions (see Fig. 2) for the insertion of Np-237 targets. The qualification process includes verifying the position-specific target configurations can safely operate under the ATR's normal and accident scenarios. Given the target design originated at ORNL for use in HFIR, the concert of components that make up the position-specific target configurations were verified for compliance. Safe performance is predominantly demonstrated to show the target has thermal margin (i.e. no melting, boiling, etc.) under various bounding conditions.

The diameters of both positions are large enough to accommodate seven targets each. The target was designed and optimized for the HFIR core, which is roughly half the length of the ATR core. As a result, spacers are used to align the center of the targets with the midplane of the ATR core to maximize Pu- 238 yield.

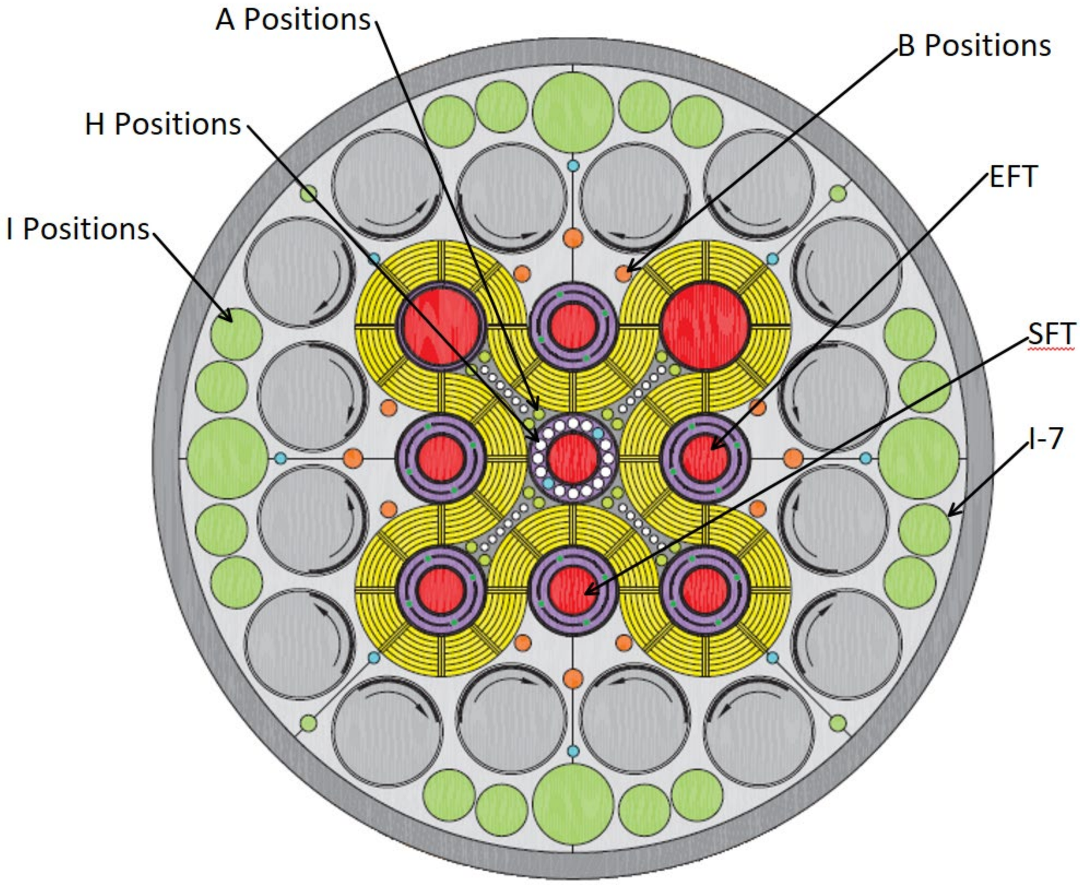


Fig. 2. Cross-sectional view of ATR.

The “I” positions are outside of the main core and have a lower thermal flux of 1 to 9 x 10^{12} n/cm^2-s compared to 4.4 x 10^{14} n/cm^2-s in flux traps [ref. 6]. Lower flux in I-positions reduces plutonium production rate, making in-core positions more attractivein terms of meeting heat source material production goals. . However, the I-positions give a higher assay of Pu-238 due to the higher ratio of thermal to fast flux, which reduces the overall likelihood of neutron interactions with the Pu-238 and fast fission fissions of Pu-238. The higher assay of material produced via I-positions is beneficial because it can be blended with old heat source material to bring it up to current specifications. To achieve sufficient production from I-positions, the targets must be irradiated for approximately six cycles. The I-positions were originally used due to their availability – I positions are less in-demand than the in-core positions due to the lower neutron flux. The in-core SFT requires only a single 60-day irradiation cycle to achieve appropriate Np to Pu conversion production. This reduction in irradiation time is beneficial for achieving higher production of Pu-238 per reactor cycle. Downsides of high flux positions include higher breeding of Pu-239 and longer cool down times due to Pu-239 fission. Future qualification efforts will be for additional in-core positions due to their higher production rate as well as I-positions due to their higher target assays.

II. OVERVIEW IRRADIATION QUALIFICATION DESCRIPTION

II.A. Overview of Design Support

The ATR contains multiple irradiation sites with variable flux magnitudes and physical size constraints, making it a versatile and flexible facility for supporting Np-237 target irradiations. To meet safety provisions, many of these core irradiation positions require initial irradiation in a low power facility known as the ATR Critical (ATR-C) Facility. ATR-C testing validates analytical models to satisfy rigorous safety requirements. These, and a myriad of other considerations, including availability and requirements for specialized tooling to handle targets down to 30-feet of water, led the project to first consider the medium I-7 position. The I-7 position has a relatively low flux outside the serpentine fuel core region and did not require preliminary ATR-C irradiation. Therefore, limited irradiation assembly and tooling design was needed to qualify the I-7 position. Later, the SFT position became available and was used with existing irradiation housing, which also reduced new hardware design and fabrication efforts. Equipment for the I-7 position is a different geometry than what will fit in the SFT irradiation position. SFT irradiations enable higher production rates than that of the I-7 position. Figure 3A shows the relative locations of these two positions. The SFT positions are partially surrounded by the serpentine fuel elements, which provides closer proximity and better exposure of this target position to the neutron flux.

Target qualification was initiated using existing target designs and, as feasible, existing hardware. Handling an axial location of the targets was accomplished by modifying common basket designs and using spacers to axially align targets to the core midplane. Figure 3B shows an image of seven production targets installed in the SFT, preparing for irradiation in ATR cycle 169A (in Spring 2021).

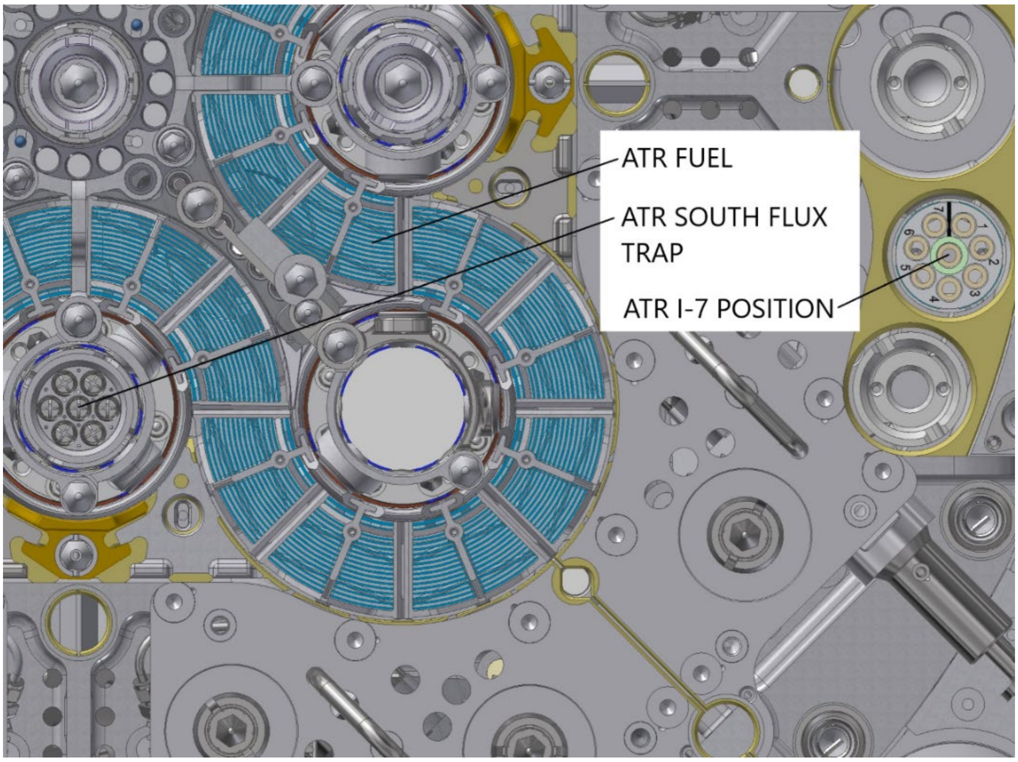


Fig. 3A. Relative Location of I-7 and South Flux Trap Positions.

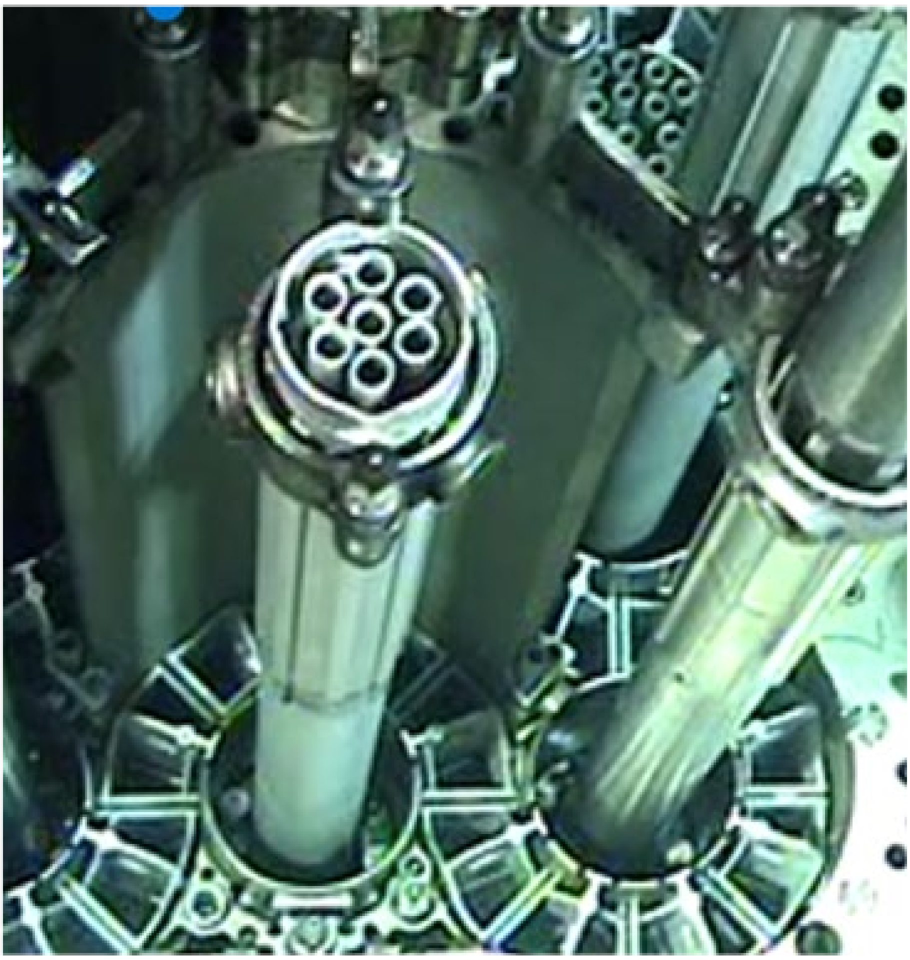


Fig. 3B. Photo, Installed South Flux Trap Assembly.

All current and future target designs intended for ATR irradiation require irradiation baskets that are unique to each type of irradiation position. The irradiation baskets and other specialized tools must be developed to carry out reactor insertion/removal, canal handling, storage, and transportation cask loading operations of the targets. Complete production and handling capabilities were developed in parallel with the initial I-7 and SFT irradiation hardware, including fabrication of a dedicated BEA Research Reactor (BRR) cask and associated payload licensing to enable shipment of irradiated targets to ORNL for post-irradiation processing.

II.B. Overview of Neutronics Qualification

Qualifying experiments for irradiation in ATR require the integration of various teams to create the appropriate models and analyses. This process begins with a neutronics analyst building a model (see Fig. 3) that reflects the computer aided design (CAD) rendering developed by a design engineer. The expected ATR operational parameters used in the model must be assumed for the model to estimate the irradiation induced heating, fission gas production within the target material, fission density within the target material, radionuclide inventory at reactor discharge, moderator temperature coefficient of reactivity for a range of water temperatures, and reactivity generated by the experiment at the beginning and end of irradiation, since ATR operational time and power varies from cycle to cycle. The assumed parameterare generally selected to bias the model in a more conservative (safer) manner because the model results yield derived parameters that demonstrate experiment compliance with the ATR safety analysis report (SAR). The Monte Carlo N Particle (MCNP) is the analysis tool used to perform these calculations. Material activation and depletion calculations are performed with the Oak Ridge Isotope Generation (ORIGEN) code and using MCNP generated neutron flux and cross section data as inputs.

A baseline requirement of a 60-day cycle was used for the Np-237 target assembly simulation. In the SFT case, the Pu-238 yield from the seven targets was estimated to be 30 grams total. This yield is slightly more than twice the amount estimated to be produced from seven targets in the I-7 position of ATR, and in one-fifth the time. Irradiation of Np-237 targets in the I-7 and SFT was completed in April 2021.

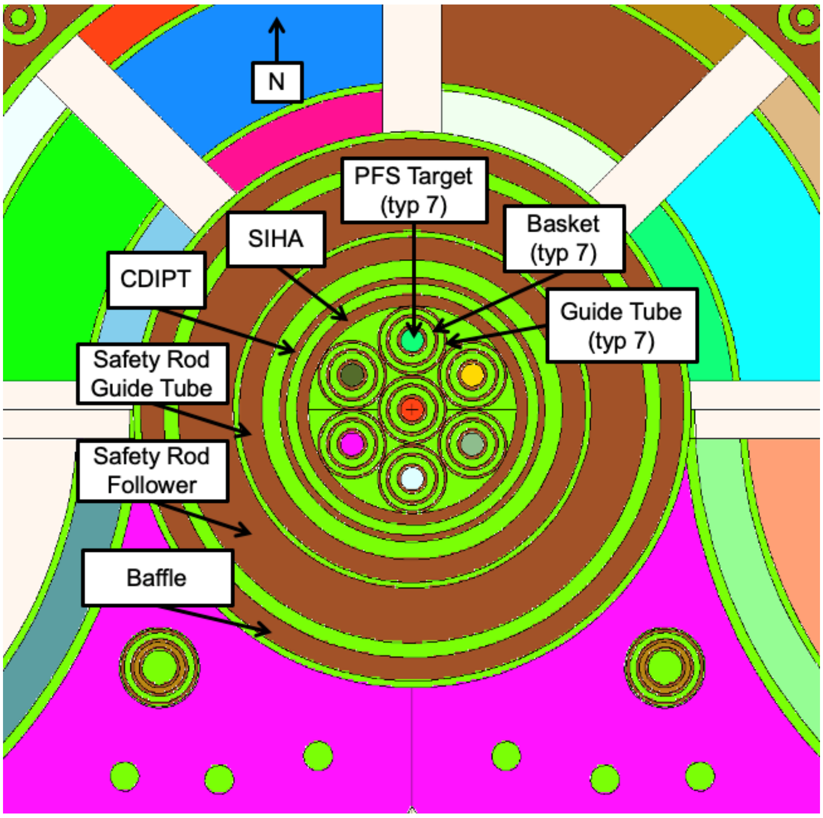


Fig. 3. Cross-sectional view of the MCNP model of the Pu-238 target assembly in ATR South Flux Trap.

II.C. Overview of Thermal Qualification

Thermal qualification was dependent on the neutronic qualification and involved several iterations to assure safety. The goal of the iterative analysis was to prevent capsule failure from overheating and to calculate the minimum required post-irradiation cooling time,. This analysis was executed using RELAP5-3D and ABAQUS. RELAP5-3D simulates thermal and hydraulic phenomena using a finite volume methodology, whereas ABAQUS employs finite element analysis.

The RELAP model (see Fig. 4) describes the hydraulic volumes through which coolant flows. Energy generated/input into the system is accounted for via heat structures attached to these flow volumes (see Fig. 5). Heat generation rates (HGRs) resultant from the neutronics analyses for fuel, structural, and coolant materials are modelled using a one-dimensional conduction model. Energy is then advected into the coolant flow volumes. A similar model was built to describe thermal and hydraulic behavior during flow stagnation and reversal.

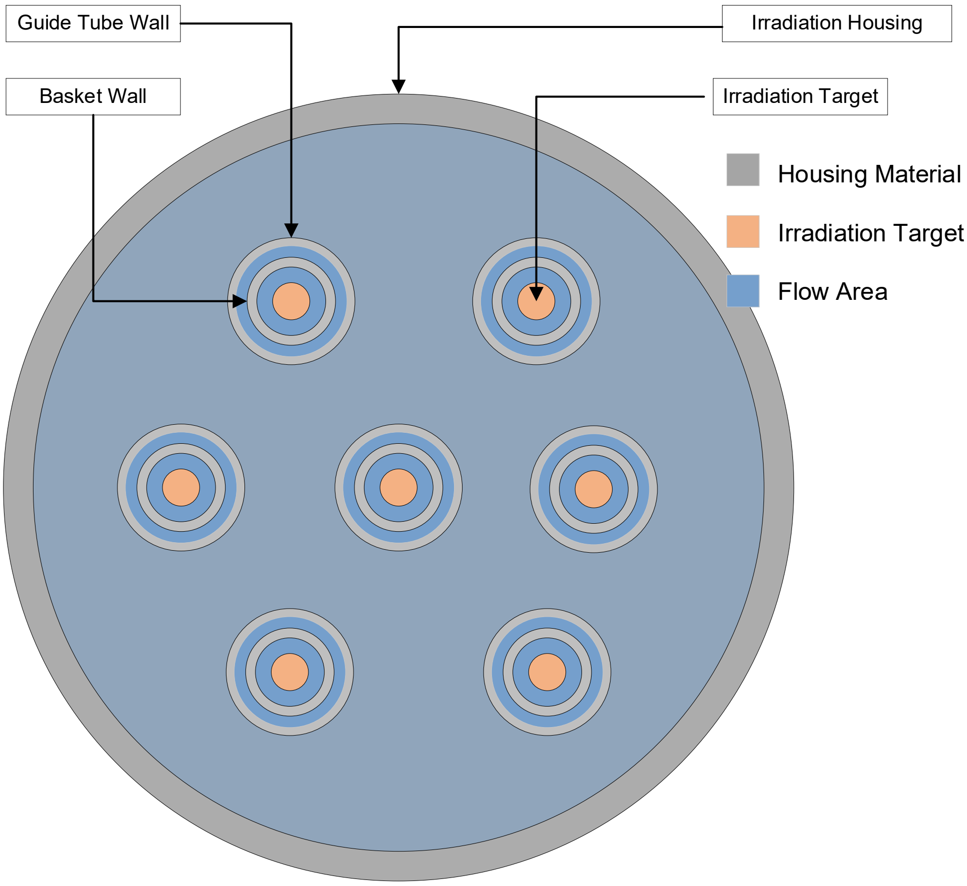


Fig. 4. Cross sectional view of the hydrodynamic system modeled using RELAP4-3D.

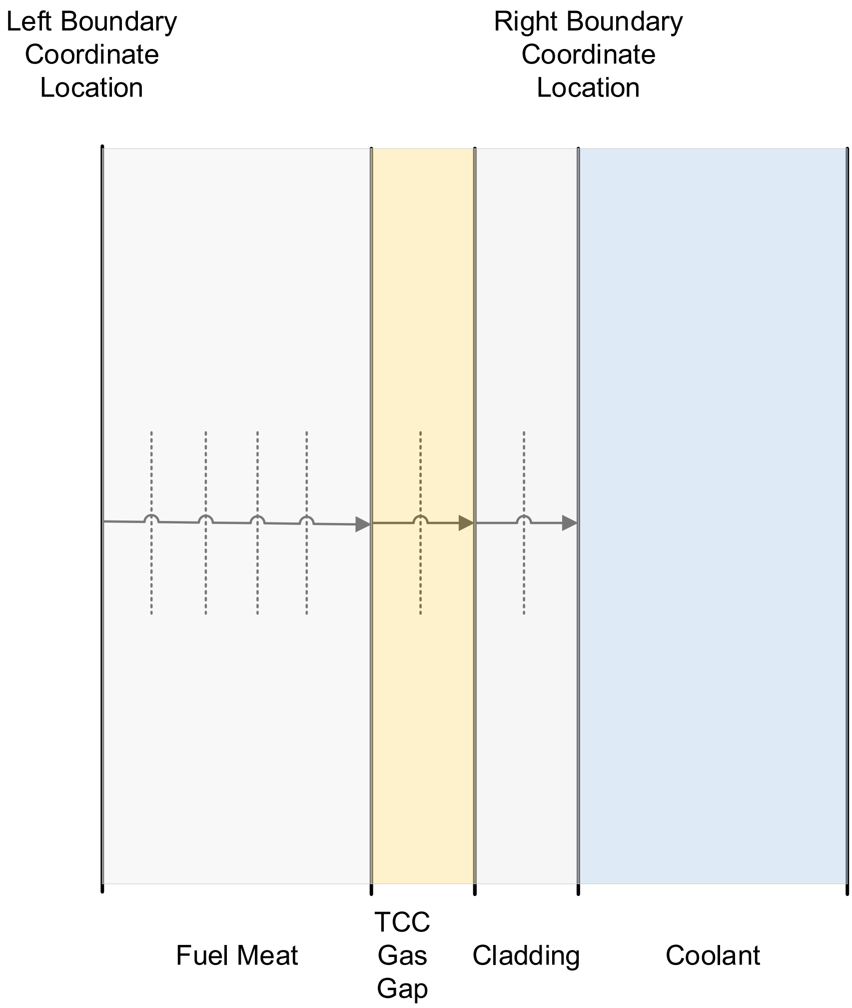


Fig. 5. Radial view of the conduction network modelled in RELAP5-3D.

Several finite-element models were developed in ABAQUS to assess the thermal performance of the Pu-238 production experiment in the SFT of the ATR. Component heat generation rates and flow conditions for each model are provided from preceding reactor physics (MCNP) and hydraulic (RELAP5-3D) analyses, respectively. Uncertainties in the modeled heating rates due to operational lobe power, instrument measurement error, and outer shim control cylinder rotation are accounted for with a safety factor multiplier. The thermal/hydraulic conditions for each model are modified to represent normal operation and possible accident scenarios in the ATR. To meet the safety requirements for operation in the ATR, the minimum departure from nucleate boiling ratio (DNBR) and flow instability ratio (FIR) for each scenario must be greater than 2. In addition, the peak temperatures of the irradiated components must remain below their respective melting points. The limiting thermal case is a reactivity insertion accident caused by a large pipe break, RIA4. The resulting temperatures from the finite-element model in the case of a RIA4 event are shown in Figure 6. The maximum temperatures of the NpO_2-Al cermet pellets and Al-6061 cladding are maintained below their respective melting points (estimated conservatively at 660°C and 585°C); while the minimum DNBR and FIR values are 2.2 and 3.9, respectively.

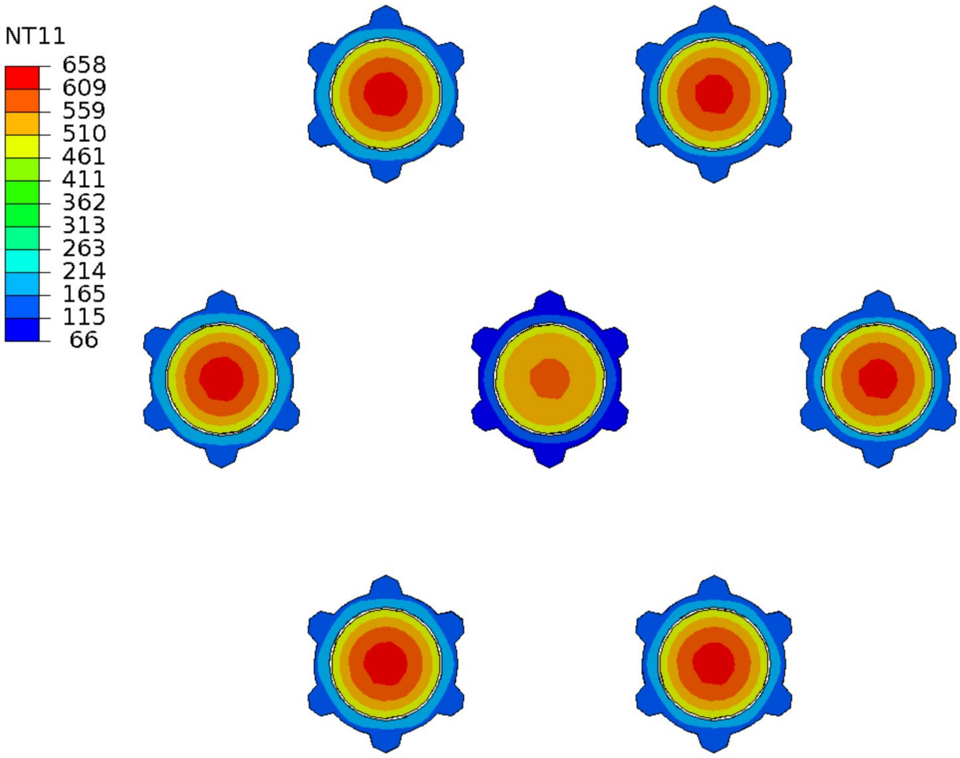


Fig. 6. Radial distribution of nodal temperature values of the NpO_2-Al cermet pellet and Al-6061 during a condition 4 reactivity insertion accident (RIA4).

II.D. Overview of Structural Qualification

The purpose of the structural safety analysis was to evaluate the target and the associated hardware under various potential loading scenarios to ensure the safety of operational personnel and the public. The 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 exerted on the target, 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. Other loadings, such as handling loads from transferring components to and from the reactor, were also considered. The decision for which loading scenarios had 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 the following: 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 extremely low probability of occurrence, and those where the loss of pressure boundary integrity meet the safety limits defined by INL's SAR, were excluded from the structural evaluation.

The response of each structural component (i.e., stress, strain, deformation) under the various loading conditions was calculated using hand calculations, or, using finite element software ABAQUS, where simplifications could not be made. These responses were compared to acceptance criteria. For the non-pressure retaining components, this criterion was typically the yield strength of the material at a given 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 applied, this code was used because it provides a nationally accepted design/analysis approach which INL has applied and adapted to various nuclear experiments. Based on the low internal pressure of the target (240 psig), the requirements of ASME Section III, Class 3 vessels were used as a guide. (ref. 7) This code defines these limits based on Design and Service Levels. Following the design specification of the ATR, which categorizes these loading scenarios into Service Levels, the response of the target resulting from each load scenario was compared to the corresponding limit in the code. Each structural component met the safety requirements and was permitted for irradiation in the ATR.

II.E. ATR-C Initial Experiment on NpO_2 and other sensors (PFS-1)

PFS-1 was a preliminary experiment that was conducted using smaller samples of NpO_2 - which were called sensors and helped to better characterize Np-237 cross sections and estimate Pu-238 production rates. These samples had a mass of 6 mg compared to 46.7g used in the ATR production targets and were irradiated in the ATR-C at a power of 600 W for 20 min compared to 150 MW for 60 days in the ATR. ATR-C is a low power duplicate of the ATR with nominal powers at 600 W vs 110 MW. Initial irradiation testing of Np-237 was carried on two NpO_2 sensors in the ATR-C reactor to estimate Pu-238 production rate. In addition to the approximately 6 mg of Np, the sensors contained copper and gold in configurations with and without cobalt sleeves Co was used because it has a high cross section for thermal neutrons.. This was done to differentiate between fast versus slow neutrons in flux profile. Considering the very low power of the ATR-C and the short irradiation time, the production of Pu-238 was only detectable due to the occurrence of the short-lived, intermediate product Np-238, which has a high gamma yield in a suitable for detection energy range. The Pu-238 production was estimated due to the presence of intermediate radioisotope Np-238, which generated characteristic gamma rays in the range 900–1,040 keV with a half-life of 2.1 days before beta-decaying to Pu-238. A comparison of the gamma-ray spectrum before irradiation (red) and after irradiation (blue), is displayed in Figure 7. The “red” spectrum shows only flat background, while the “blue” spectrum has a presence of several peaks after irradiation.

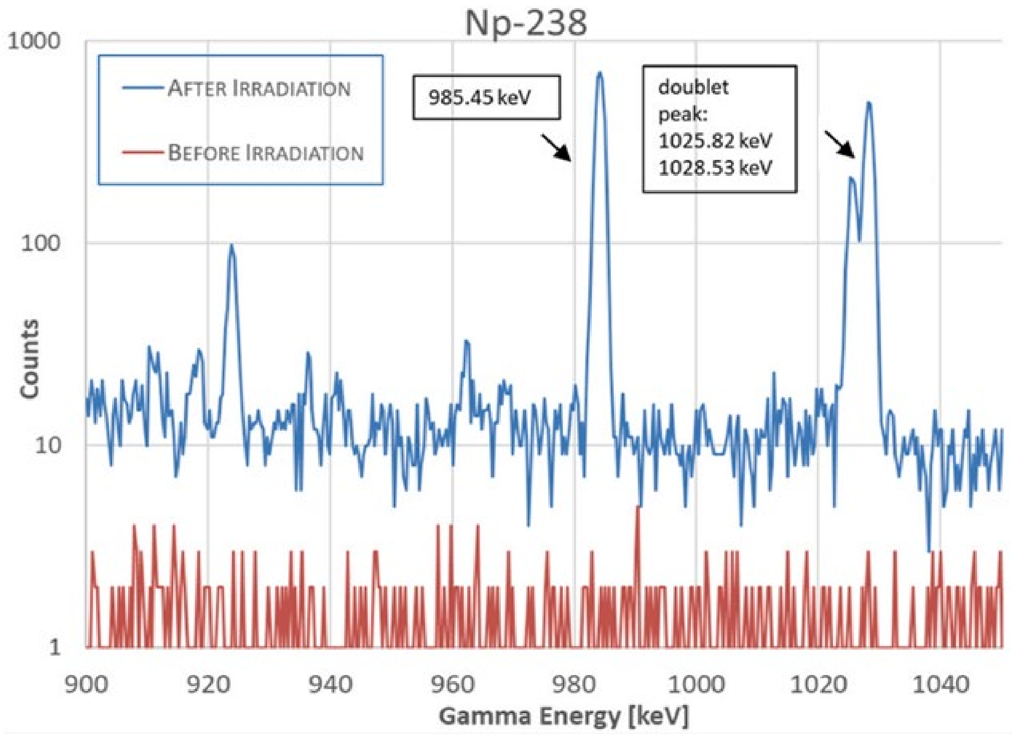


Fig. 7. Gamma spectrum from neptunium dioxide sensor.

The ATR-C irradiation lasted only 20 minutes (compared to 60 days for the ATR cycle). The irradiation had to be followed by approximately 2 hours of cooling time due to the presence of short-lived radioisotopes, mainly from the Aluminium clad and matrix. The measurements of these gamma rays were carried out in short and long time-ranges: for approximately 2 hours, and then for 1-2 days until up to 3.5 days later. This allowed not only to determine the Np-239 production rate with better certainty, but also to verify the slow/fast flux effective rate due to Cu/Au ratio. Isotopes Au-197 and Cu-63 have different cross sections for gamma capture in thermal, epithermal, and fast ranges, they also have different decay times, therefore, carrying short and long times helps to verify neutron flux characteristics. The ATR-C experiment showed in the I position a production rate at 10% higher than predicted from modelling at 1.01×10^{-2} µCi/mg. This value is in agreement with an earlier trade study that predicted assay value

(Pu-238/total Pu) as high as 98% for "I" positions after one year and the measurement gave certainty that predicted production rates at the ATR could be achieved at the large scale [ref. 2].

II.F. SAFETY ANALYSIS AND DOCUMENTATION

An experiment safety analysis (ESA) was developed for the Pu-238 production experiment in both the I-7 positions and in the SFT positions. Both ESAs demonstrate the Pu-238 production experiments' irradiation in the ATR are in compliance with the requirements of technical safety requirements and the approved authorization basis established by ATR's SAR. The ESAs were developed and authorized under an ATR Complex procedure that addresses experiment receipt, reactor loading, irradiation, discharge, storage, preparing for shipping from ATR, and waste disposal. The Pu-238 production ESAs concluded that operation of the Pu-238 production experiments were in accordance with the restrictions identified in the ESAs and within the authorization basis of the ATR.

III. OPERATIONS

PFS-3 targets were shipped from ORNL to the INL ATR Complex individually in shipping drums. The targets were then unloaded from the shipping drums and taken to the ATR-C facility for preliminary irradiation. Aspects of this activity are shown in Figure 8. The FPS-3 targets were the first production targets irradiated in ATR and ATR-C. Previously, PFS-1 flux wires were irradiated in ATR-C to confirm calculated cross sections and estimate full scale production yield, and the PFS-2 experiment was planned as a way to perform dosimetry in the canter bail of the I-7 position. Work on PFS-2 was stopped when initial design reviews determined it was not necessary. Pu-238 production targets were irradiated at extremely low power in the ATR-C facility to ensure safety of the targets prior to full power irradiation in ATR and is shown in Fig 9. Irradiation in the ATR-C facility measures the reactivity worth of the Pu-238 production targets and helps provide characterization and projections of how the targets will affect the ATR core power distribution during irradiation. This process is typical for 'fuelled' experiments, those containing fissile material and is necessary to ensure that the insertion of the experiment does not compromise the safe operation of the ATR.

Fig. 8. ATR Personnel remove a PFS-3 production target from the shipping drum while monitoring radiation levels.

Fig. 9. ATR-C facility personnel remove and arrange irradiated flux wires for analysis.

After completion of the ATR-C test run, the targets were transferred to the ATR canal for configuration prior to insertion in ATR (see Figure 10). The assembly was then transferred under water by ATR Canal Operators to the ATR drop chute. ATR reactor top operators were able to retrieve the assembly from the inner vessel side of the drop chute. The Pu-238 production target assembly was inserted into the South Flux Trap and inventoried to ensure the assembly was properly seated into the chopped dummy in-pile tube.

Fig. 10. ATR Canal Operators load a basket containing a Pu-238 production target into the South Irradiation Housing Assembly.

IV. FUTURE WORK

The INL team is currently working to qualify several ATR positions, as detailed in Table 1, with an updated target design called the ATR Gen 1 target. This new target design will utilize the full length of the ATR core and will be comprised of two targets stacked on top of each other. Having two shorter top and bottom targets is desirable because it allows common processing methods to separate the Pu-238 from the post-irradiated targets in the hot cells at ORNL.

INL plans to initially qualify the Northeast Flux Trap (NEFT), Inner A positions, and H positions with the ATR Gen 1 target design. The PFS-3 target design was only about half the height of the ATR core, so using the ATR Gen 1 targets will increase production per ATR irradiation position as compared to the PFS-3 targets. Later, the B positions, SFT, and East Flux Trap will be qualified with ATR Gen 1 target design. The low flux in the I-positions results in a low production rate, which makes them the lowest priority position to qualify while higher producing inner core positions are available.

TABLE I. ATR Locations and Number of Positions.

Column Header	Positions in Target	# of Locations in ATR
NEFT	23	1
Inner A	1	8
H Position	1	14
B Position	1	8
South Flux Trap	7	1
East Flux Trap	7	1
I Position	1 to 7	23

V. CONCLUSIONS

Idaho National Laboratory's work on Pu-238 production in ATR has qualified the SFT for ORNL supplied targets and is ramping up the production process to meet a 0.5 kg of heat source material produced per year by 2025. Seven targets were characterized by an ATR-C run and inserted into the SFT for irradiation in ATR cycle 169A. These targets, as well as several targets being irradiated in the ATR I-7 position, are expected to produce several 10s of grams of Pu-238. These targets will be sent to ORNL for post-irradiation processing in 2022. The recovered Pu-238 will ultimately be used as fuel for Radioisotope Power Systems in future space exploration missions.

ACRONYMS

ASME - American Society of Mechanical Engineers

ATR - Advanced Test Reactor

ATR-C - Advanced Test Reactor Critical

BEA - Battelle Energy Alliance

B&PV - Boiler and Pressure Vessel Code

BRR - BEA Research Reactor

CAD - Computer Aided Design

DNBR - Departure from Nucleate Boiling

DOE - Department of Energy

ESA - Experiment Safety Analysis

HFIR - High Flux Isotope Reactor

INL - Idaho National Laboratory

MCNP - Monte Carlo N Particle

NASA – National Aeronautics and Space Administration

NEFT - North East Flux Trap

Np -Neptunium

NpO_2-Al – Neptunium Oxide-Aluminum

ORIGEN - Oak Ridge Isotope GENeration

ORNL - Oak Ridge National Laboratory

Pu - Plutonium

RIA4 - Reactivity Insertion Accident, Condition 4

RPS – Radioisotope Power Systems

SAR - Safety Analysis Report

SFT - South Flux Trap

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