



## THE EVOLUTION OF HFIR CERMET PU-238 PRODUCTION TARGETS

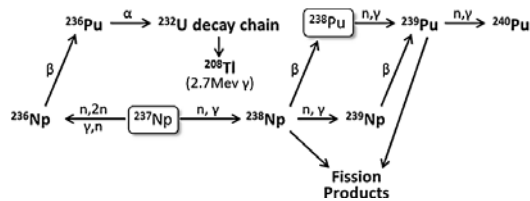
Richard Howard<sup>1</sup>

<sup>1</sup>Oak Ridge National Laboratory, Box 2008 MS6051, Knoxville, TN, 37931-6051, 865-576-4867, [howardrh@ornl.gov](mailto:howardrh@ornl.gov)

The production of  $^{238}\text{Pu}$  as a heat generation source for radioisotope thermoelectric generators (RTGs) was active at the Savannah River Site (SRS) from the 1960s-1990s. Stockpiles of this valuable radioisotope have decreased over the last few decades and more material must be produced to support future deep space exploration missions. Oak Ridge National Laboratory (ORNL) has initiated efforts to establish a reliable production facility to replenish this resource by transmuting  $^{237}\text{Np}$  into  $^{238}\text{Pu}$ . Like the SRS, ORNL uses a mechanically pressed ceramic/metallic (cermet) pellet that blends  $^{237}\text{Np}$  (in the form of ceramic  $\text{NpO}_2$  powder) with aluminum powder. A “Generation I” target containing 52 such pellets has been qualified for irradiation at the High Flux Isotope Reactor at ORNL. The next “Generation II” target design incorporates lessons learned from the previous qualification exercise to develop a further optimized irradiation vehicle design. This paper discusses the evolution of the cermet targets at ORNL and provides details on the Generation II design.

### I. BACKGROUND

The U.S. Department of Energy (DOE) supplies radioisotope power systems, generally radioisotope thermoelectric generators, to the National Aeronautics and Space Administration (NASA) for use in deep space mission activities. Plutonium-238 ( $^{238}\text{Pu}$ ) oxide pellets provide the heat source for these power systems via alpha decay, with a half-life of 87.7 years. This decay heat is used to generate electrical power via thermoelectric conversion and ambient heat for NASA instruments and space vehicles. Oak Ridge National Laboratory (ORNL), which is a member of the DOE laboratory system, is managing ongoing efforts to re-establish a  $^{238}\text{Pu}$  production facility to replenish the US stockpile of this valuable radioisotope (Ref. 1). The current workflow irradiates  $^{237}\text{Np}$  in the form of  $\text{NpO}_2$  at the High Flux Isotope Reactor (HFIR), to produce  $^{238}\text{Pu}$ . The neutron transmutation path for this process is shown in Fig. 1.



**Fig. 1.** Transmutation pathway for  $^{237}\text{Np}$  to produce  $^{238}\text{Pu}$  (Ref. 2).

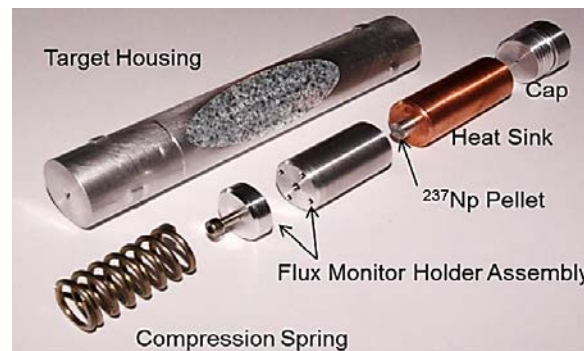
Following neutron irradiation, the radioisotope is collected through a complex radiochemical separation process that tends to produce high volumes of high-activity radioactive waste (Ref. 2). Additionally, radioactive solid components not eliminated through some radiochemical means require a disposal plan. Processing all this material is expensive, and waste production will increase proportionally with the planned growth in  $^{238}\text{Pu}$  production. Therefore, it is prudent to design production targets that maximize  $^{238}\text{Pu}$ , while minimizing the waste stream.

Furthermore, the target design must be robust and perform adequately under irradiation conditions to ensure a reliable production process can be established to support this mission. This is demonstrated through performing destructive and non-destructive testing, experimentation, and multiphysics analyses which provides a safety basis for the target design. Details of the design improvements and validation methods are described in this work.

### II. EARLIER TARGET CONCEPTS AND GENERATION I TARGET DESIGN

#### II.A. The Single Pellet Target

The initial target design produced at ORNL was called the single pellet target. The focus of this target was to provide post-irradiation examination (PIE) data for the cermet pellet and irradiation performance data for subsequent, multi-pellet irradiations. As the name suggests, the target housed a single aluminum matrix/ $\text{NpO}_2$  cermet pellet target. The assembly configuration of the single pellet target can be seen in Fig. 2. More details on this particular experiment and subsequent performance results can be found elsewhere (Ref. 3, 4).



**Fig. 2.** Part config. of the single pellet target (Ref. 3).

## II.B. Generation I Target

The next phase in target development saw the deployment of the Generation I, or gen-I, target. This design started as a partially loaded target containing eight cermet  $\text{NpO}_2$  pellets. The containment consisted of an Al-6061 (T-4 temper) finned extrusion tube with aluminum alloy end caps. The overall length of the target was roughly 84 cm, with the pellet stack comprising roughly 5 cm. This implies that a majority of the target was void space, which provided excess volume for the unknown fraction of fission gas release. PIE from the single pellet experiments indicated the pellets see a net shrinkage during the irradiation cycle. Therefore, the extrusion housing was hydrostatically collapsed onto the pellets to reduce the radial gaps between the pellet and housing during irradiation. The unsupported sections of the target were supported internally with stainless steel reinforcement tubes to ensure the target containment remained intact during the collapse process.

These partially loaded targets provided multi-cycle irradiation performance data to support subsequent fully loaded targets. PIE data gathered from this experiment set informed neutronics and heat transfer models to establish a conservative fully loaded target safety basis for a single HFIR irradiation cycle. The irradiation program then shifted to solely producing and irradiating fully loaded targets for one cycle. This phase provided un-irradiated nondestructive, destructive, and PIE data to relax this conservatism and inform the multi-cycle gen-I target design that is currently in use for  $^{238}\text{Pu}$  production at ORNL. Hurt, et. al. provides a comprehensive study on this evolution, and specifically describes heat transfer modelling of the gen-I target using COMSOL (Ref. 3). A schematic of the current gen-I fully loaded target can be seen in Fig. 3 (not to scale).

The gen-I target has been extensively overdesigned to maintain the integrity of the containment. Components such as the expansion assembly (exp. assem. as seen in Fig. 3) were intended to eliminate axial loading of the target. Other features that provide an engineered rupture relief, in the event of an over-pressurization event, were also employed. Over time, extensive destructive testing

was performed on cermet pellets and prototypic target containments to quantify performance limitations of each of these components, and weigh the usefulness of the extraneous safety features.

The original safety basis for the gen-I target design was written to take credit for these features, and eliminating those that were found to be superfluous was not trivial. Furthermore, the target's final hermetic closure relied on a manual tungsten inert gas (TIG) aluminum weld, which proved to be both difficult and failure prone. Lastly, the target holder, used to install a set of seven gen-I targets into the HFIR, was designed for single use and was expensive to fabricate and dispose. All these factors lead to costly and difficult fabrication campaigns. To work towards establishing a large scale  $^{238}\text{Pu}$  production facility, these challenges had to be overcome to ensure success of the mission. Therefore, further development of the gen-I design was discontinued, and a separate Generation II (gen-II) target design effort was initiated.

## II.C. Generation II Target

Design improvements for the gen-II targets include:

- Elimination of the aluminum closure weld(s),
- Reduction or elimination of extraneous parts and design features
- Development of a new holder design that allows for recycling of parts

These improvements are constrained such that they cannot negatively impact the cermet pellet performance, which includes  $^{238}\text{Pu}$  production rates, thermal boundary conditions, target containment integrity, and several other criteria. Therefore, the gen-II mimics the active length of the gen-I target, using the finned extrusion that is hydrostatically compressed onto the pellets prior to irradiation. Other ancillary parts/features, such as the dummy pellet and weld joints were modified or abandoned. For example, it was ascertained that the external compression operation locked each of the internal components into an individual pocket within the target. As such, local axial stress applied by the pellet in

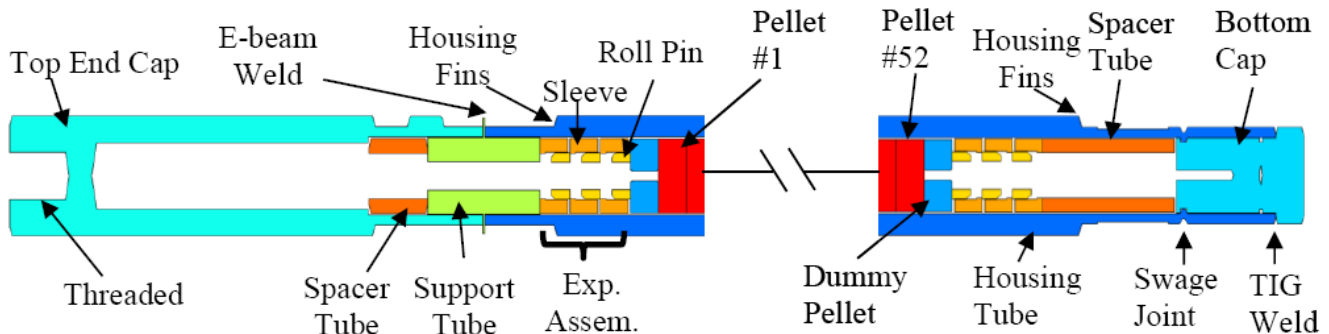
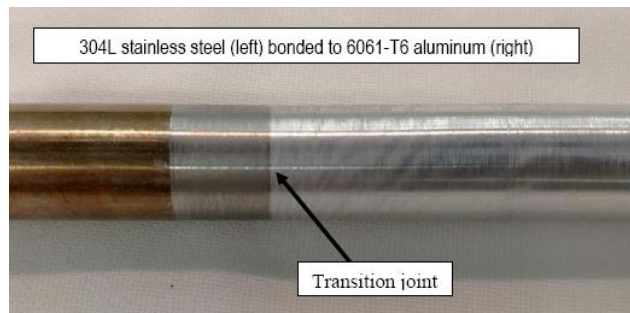


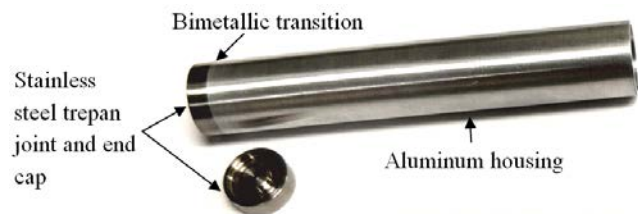
Fig. 3. Schematic of the gen-I target (Ref. 3).

the finned region to the target containment did not affect the structural integrity of the housing. Moreover, PIE did not reveal any indication that the expansion assemblies were being compressed. Therefore, this feature was abandoned in the gen-II target.

Other internal parts were modified slightly to reduce complexity of fabrication, but the most significant potential changes pertain to the welded components. As stated earlier, relying on a challenging aluminum closure weld is dubious (Ref. 5). Moreover, performing this weld in a glovebox environment adds an additional set of obstacles. Therefore, bimetallic stainless steel-to-aluminum transition joints (Fig. 4) could be incorporated into the target design to simplify welding. Note that the brassy colored section of the transition joint seen in Fig. 4 is un-machined stainless steel. In contrast to aluminum alloys, it has been shown that stainless steel alloys generally have good weldability under a wide range of conditions (Ref. 6). The usage of bimetallic transition joints to improve fabrication or performance of irradiation capsules is not a new concept (Ref. 7). Moreover, researchers are currently working to qualify bimetallic weld joints to be used for the general HFIR irradiation capsules, referred to as rabbits (Fig. 5). This new rabbit format would simplify capsule fabrication in hot cells, which is directly aligned with the mission of the gen-II design, which is to improve the reliability of target production.



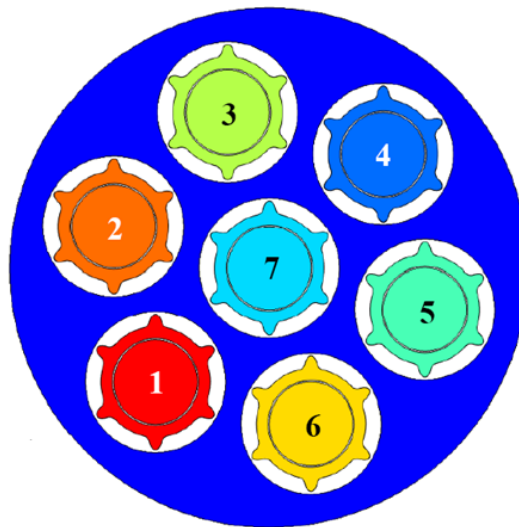
**Fig. 4.** Bimetallic transition joint.



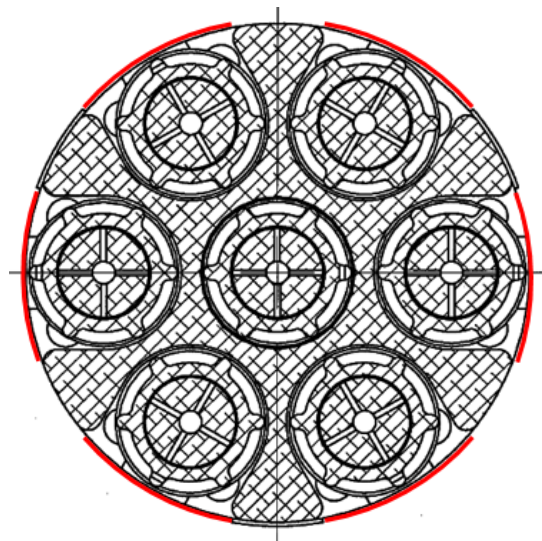
**Fig. 5.** Usage of a bimetallic transition joint in a rabbit.

The target holder was also redesigned to provide ease of loading and to allow for some recyclability of parts, reducing high-activity waste. The original holder was fabricated from a solid round bar aluminum extrusion with seven holes running in the axial direction. See Fig. 6

for a graphical representation of the radial cross-section of the holder. Although it is a simple configuration, it does not lend itself to easily load targets. The external compression operation tends to mildly distort the straightness of the targets, imparting a long axial arc onto the assembly. In order to mitigate this effect, an open channel “spider” extrusion was developed. Like its predecessor, it can house seven targets, but only has a single tubular channel in the center. The remaining target sites have an open perimeter so that the targets can be loaded from the radial direction, as opposed to being slid into place along the axial direction of the extrusion. A radial cross-section of the gen-II spider extrusion can be seen in Fig. 7.



**Fig. 6.** Radial cross-section of the gen-I target holder (Ref. 3).



**Fig. 7.** Radial cross-section of the gen-II spider extrusion (open perimeter in red).

A great deal of testing, experimentation, and thermal-hydraulic analyses were performed to demonstrate the performance equivalence between the gen-I and gen-II target configurations. Primarily, a set of qualification tests and acceptance criteria were established for the bimetallic transition joint. Similarly, flow experiments were performed to inform the spider extrusion design and validate thermal-hydraulic safety basis calculations. These efforts were able to demonstrate equivalency between the two configurations and establish a sound safety basis for the gen-II target configuration (Ref. 7).

### III. FUTURE WORK

The gen-II target format is currently being formally qualified for use at the HFIR. This includes safety screening and acceptance of the safety basis documentation developed for this design. A total of 63 gen-II targets are commissioned for insertion in FY18. Fabrication and assembly of these targets are currently ongoing to support this schedule.

Lessons learned from the initial fabrication and assembly process of these first targets are being incorporated to further optimize the gen-II target design. This includes further simplification of internal components and developing more robust weld joints. The current design utilizes manual TIG aluminum welds to attach the bimetallic transition joints to the extrusions, but this process is challenging and may not be scalable for production volumes. Therefore, a more stable and repeatable electron beam weld joint is being developed to replace the manual TIG process.

The initial set of gen-II targets are planned for insertion in the third quarter of FY-18.

### ACKNOWLEDGMENTS

Funding for this program is provided by the National Aeronautics and Space Administration through the U.S. Department of Energy, Office of Nuclear Energy.

This manuscript has been authored by UT-Battelle, LLC under Contract No. DE-AC05-00OR22725 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes. The Department of Energy will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan.

### REFERENCES

1. R. WHAM, L. FELKER, E. COLLINS, D. BENKER, R. OWENS, R. HOBBS, D. CHANDLER and R. J. VEDDER, "Reestablishing the Supply of Plutonium-238," in *American Institute of Aeronautics and Astronautics Propulsion Energy Conference*, Orlando, FL, 2015.
2. E. COLLINS and R. WHAM, "Development of Improved Targets, Separation Processes, and Waste Management for Pu-238 Production" in *International Congress on Advances in Nuclear Power*, San Francisco, CA, 2016.
3. C. HURT, J. FREELS, R. HOBBS, P. JAIN and G. MALDONADO, "Thermal Safety Analyses for the Production of Plutonium-238 at the High Flux Isotope Reactor," Oak Ridge National Laboratory, ORNL/TM-2016/234, Oak Ridge, TN, 2016.
4. C. HURT, R. WHAM, R. HOBBS, R. OWENS, D. CHANDLER, J. FREELS and G. MALDONADO, "Plutonium-238 Production Target Design Studies," in *INMM 55th Annual Meeting*, Atlanta, GA, 2014.
5. G. MATHERS, *Welding of Aluminium and Its Alloys*, Cambridge Woodhead Publishing Limited, 2002.
6. P. KORINKO and S. MALENE, "Considerations for the weldability of types 304L and 316L stainless steel," *Practical Failure Analysis*, vol. 1, no. 4, 2001.
7. R. HOWARD, J. McDUFFEE, Y. KATOH "Graphite Compressive Creep Capsule Design for Irradiation in the HFIR," in *2013 ANS Annual Meeting*, Atlanta, GA, 2013.