

AUTOMATION OF NEPTUNIUM OXIDE-ALUMINUM TARGET FABRICATION

Robert M. Wham¹, R. Steve Owens, Raymond James Vedder, James Henry Miller, Scott Pierce²

¹Oak Ridge National Laboratory, 1 Bethel Valley Road, Oak Ridge, TN, 37831, 865-241-0873, whamrm@ornl.gov
²Trident Industries LLC, 9222 Tedford Road, Knoxville, TN 37922, 865-228-9758

Reestablishing the supply of plutonium-238 dioxide has required design and development of new targets for irradiation to convert neptunium-237 to plutonium-238. Test target irradiations have defined appropriate operating conditions for irradiation. To date target fabrication efforts have relied on manual operations for all phases of target fabrication. This paper describes new methods implemented to automate the target fabrication process and scale up from ~60 targets/year to ~200 targets/year.

I. INTRODUCTION

A critical part of the effort to reestablish a domestic supply of plutonium-238 is the fabrication of target assemblies. The targets, typically made from neptunium-237 dioxide (²³⁷NpO₂) blended with a nonreactive metal (such as aluminum), are irradiated in a reactor with a sufficiently high neutron flux and irradiation time to produce ²³⁸Pu at the needed rate. The neptunium undergoes the neutron capture reaction in Fig. 1.

 ^{238}Pu will undergo additional neutron capture to produce heavier isotopes ($^{239}\text{Pu},~^{240}\text{Pu}$). Irradiation time is adjusted to ensure that the ^{238}Pu content expressed as a fraction if the total Pu ($^{238}\text{Pu}+^{239}\text{Pu}+^{408}\text{Pu}$) remains greater than 0.85.

$$^{237}\text{Np} \xrightarrow{\text{(n, }\gamma)}$$
 $^{238}\text{Np} \xrightarrow{\text{beta decay - 2.1 d}}$ ^{238}Pu

Fig. 1. Neptunium-237 transmutation to plutonium-238.

Fabrication of the neptunium targets occurs at the Radiochemical Engineering Development Center (REDC) at Oak Ridge National Laboratory (ORNL). The targets are irradiated either in the ORNL High Flux Isotope Reactor (HFIR) or the Idaho National Laboratory Advanced Test Reactor (ATR). Figure 2 depicts the simplified steady-state processing scheme.

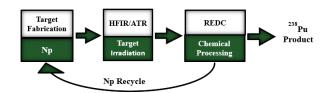


Fig. 2. Plutonium production process.

To attain the desired annual production of 1.5 kg of 238 Pu oxide, it is necessary to irradiate $^{-12}$ kg of 237 Np annually. Most ($^{-85}$ %) of the target feedstock NpO $_2$ material will be recycled from the unconverted NpO $_2$. Make up material ($^{-15}$ %) will be shipped as needed from Idaho National Laboratory to the REDC facility or to another ORNL staging area. Upon receipt of makeup NpO $_2$, the neptunium oxide feed will be dissolved, purified, then transferred to the Target Fabrication Laboratory to convert back to oxide for pellet pressing.

II. CERMET TARGET FABRICATION

The baseline irradiation target consists of a stack of cylindrical pellets containing the ²³⁷NpO₂/Al cermet target material sealed within an aluminum tube (clad). The ²³⁷NpO₂ is blended with aluminum powder and formed into pellets by pressing the material in a die at high pressures. The pressing of the oxide target material with the substrate aluminum base material forms a ceramic-metallic component typically referred to as a cermet. The cermet pellets are loaded into the aluminum tubes. and the ends of the tubes are welded to form a single target assembly. The target tube is then compressed around the pellet using hydrostatic compression. This compression decreases the gap between pellets and the outer clad. targets are placed into target holders to form target arrays, with the number of targets per array dependent upon the reactor and reactor position (Fig. 3).

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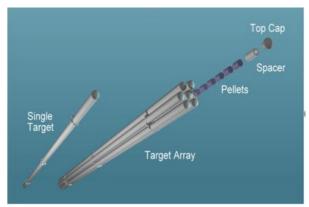


Fig. 3. Target

The pellets used in the targets are nominally 0.25 in. in diameter by 0.375 in. long as depicted in Figure 4. These pellets are fabricated by blending a mixture that contains 10-30 vol % NpO $_2$ with the balance being aluminum at 90% of the theoretical density of the blend. Pellets are currently fabricated at 20 vol % NpO $_2$. To assist with the pressing operation, it is necessary to use a lubricant in the die. After pressing, the pellets are heated to $300-500^{\circ}\text{C}$ to remove the binder/lubricant. Figure 5 is functional diagram of cermet target fabrication.



Fig. 4. Typical cermet pellet.

Functional Flowsheet for Pellet Fabrication

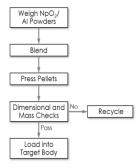


Fig. 5. Functional requirements for target

The next step in target fabrication is dimensional and mass checks to ensure the appropriate

quantities of NpO $_2$ and Al are in the pellet at the desired volume fractions. Pellet passing dimensional and mass checks are loaded into target bodies, backfilled with helium, and seal welded. Because two irradiation facilities may be used to produce 238 Pu, the active region of the target varies from ~ 20 in. in HFIR to ~ 48 in. in ATR. For this reason, it is necessary to assemble targets of varying lengths.

III. TARGET QUALITY CONTROL REQUIREMENTS

Targets will be qualified for irradiation according to guidelines in the reactor safety basis documents. The approved documents will then form a set of requirements (e.g., neptunium mass loading, materials of construction) that will establish the minimum conditions necessary to ensure that these targets can be irradiated in either HFIR or ATR. Table 1 shows the anticipated set of requirements based on previous target qualification experience.

Table 1. Quality control requirements to ensure target qualifications

Objective	Measurement	Control
Ensure target does not melt during irradiation	Uniform loading of NpO2 Aluminum phase is continuous Clad-to-pellet gap < 0.75 mil	Mass of NpO2 in each pellet Pellet collimated gamma scan Aluminum powder size Swage targets and inspect with real-time radiography
Ensure target does not leak	Helium leak test Weld inspection	 Pass helium leak test at 10⁻⁷ cm^{3/s} Real-time radiography
Ensure target survives at reactor pressure	Hydrostatic test at 1035 psi	Accept or reject after testing
Ensure proper materials of construction	Mill certification on aluminum powder and clad Neptunium and associated impurities	Vendor-supplied data Inductively Coupled Plasma/Mass Spectrometer ICP/MS

A critical aspect of the target assembly is a series of quality control checks to be performed throughout the fabrication process. From the analysis of feedstock materials through final leak testing and visual inspection, the goal of the quality controls is to ensure that every aspect of the assembly meets the requirements analyzed and credited for irradiation in the reactor (ATR or HFIR).

The following basic quality checks are performed during target assembly:

- Analysis and documentation of feedstock materials
- Pellet dimensional checks and weights
- Collimated gamma scan of pellet to check homogeneity

- Leak test of target welds
- Full-length radiography of target to check weld area and verification of loading and proper compression (swaging)
- Final visual inspection to check for defects or damage during assembly

Records of the various quality assurance checks performed during assembly will be retained in a database report that is generated with each target.

IV. AUTOMATION OF KEY TARGET FABRICATION STEPS

Target fabrication steps are shown in Fig. 4. The basic steps are as follows: (1) weigh aluminum powder into a glass vial; (2) transfer vial into glovebox; (3) weigh NpO_2 into vial; (4) mix aluminum powder with NpO_2 ; (5) transfer into a die; (6) press powder mixture into a solid pellet; (7) measure length, diameter, and weight of pellet to verify that the pellet meets design criteria. Those steps were performed by hand until October 2018. In October 2018, several pieces of equipment used to automate the process were installed and tested in gloveboxes in the Target Fabrication Laboratory at REDC. The following subsections discuss the various steps shown in Fig. 5.

- A. Weigh NpO₂/Al powders—A commercially available Mettler Toledo scale has been paired with an automated dosing system and 30 vial carousel system manufactured by Mettler Toledo. Commercial software sold by Mettler Toledo is used to manage information on NpO₂ and Al powder weights. All data on pellet weights is collected into a database for subsequent use.
- B. Blend—The vials containing Al powder and NpO₂ powder are loaded into a tumbler and mixed. The tumbler unit was scaled up to hold 30 vials.
- C. Press pellets—A specially designed automated press was built and tested. The press unit takes the glass vials, scans a barcode used to identify the pellet, removes the lid from the vial, dispenses the powder into a die, presses the powder into a solid pellet, and then removes the pellet from the die. The press also lubricates the die before pressing and cleans the die after the pellet has been ejected.
- D. Dimensional and mass checks—Off-the-shelf laser micrometers and a scale are used to automatically measure pellet diameter, length, and weight. At the end of the measurement, the system calculates the density of the NpO₂ in the matrix to ensure the volume loading is less than

or equal to 20 vol%. If the pellet meets the dimensional and weight limits, it is passed to target fabrication. If not, the pellet is then held as a reject pellet.

Overall, the pellet pressing operation has been scaled up to ~90 pellets/day versus ~30 pellets/day for the manual pressing. Other benefits of the automated system are a reduction in overall radiation dose for the workers and computer-based databases for data storage (as opposed to manual record keeping). During the operation of the automated dispensing system, there were several instances where characteristics of the oxide powder changed (increased density) which resulted in dispending errors. Studies are underway to evaluate impacts of changes in NpO₂ powder characteristics on error reduction.

V. CONCLUSION

The target fabrication rate has been scaled up from ~60 targets/year to ~200 targets/year by converting to automated dispensing and cermet pellet pressing using as many commercial-off-the-shelf items as practical. A major step in the automation process was design, fabrication, and operation of the automated pellet press representing about half of the scale necessary for full-scale production. Additional scale up is expected in FY2020.

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