Pacific Northwest NATIONAL LABORATORY

PNNL-30625

Fabrication and Characterization of Plutonium Targets for Irradiation in the Flattop Critical Assembly

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LM Arrigo
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U.S. DEPARTMENT OF ENERGY

Prepared for the U.S. Department of Energy
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Pacific Northwest National Laboratory
Richland, Washington 99354

Summary

In FY16, Pu targets were fabricated in support of the Nuclear Physics: Cumulative Fission Product Yields (Fission R-values) task of the F2016 venture project; these targets are suitable for irradiation in the Flattop critical assembly at the Nevada National Security Site (NNSS). This report documents an overview of the target design, preparation of the Pu metal materials by removing the oxide layer and appropriate sizing for the target holders, target fabrication including filling an inner aluminum cup with the Pu material and the sealing of an outer stainless steel capsule, target assay using gamma spectroscopy, and the He leak testing of the final targets. Four targets were fabricated with two containing 99% ^{239}Pu metal and two containing 93% ^{239}Pu metal. Additional samples of each material were set aside for mass spectrometric analysis at PNNL and Los Alamos National Laboratory.

Acknowledgments

The authors would like to acknowledge and thank the staff who assisted with the preparation of the Pu targets.

- The Pu materials were identified and provided by Lawrence Greenwood and Bruce McNamara.
- Preparation of the Pu materials and removal of the oxide layer was performed by Lanèe Snow and Bruce McNamara.
- Loading of the Al cups was performed by Lanèe Snow along with oversight of fabrication of the Al cups, design of the glove bags, and other laboratory activities.
- Development of the settings for the orbital welder and the target welding were performed by Michael Dahl and Stanley Pitman.
- The GEA analyses and reporting were performed by Lawrence Greenwood, Mike Cantaloub, and Truc Trang-Le.
- The He leak testing and reporting were performed by James Peterson and Crystal Rutherford.

Acronyms and Abbreviations

CA	Contamination Area
FSR	Field Services Representative
GEA	Gamma Energy Analysis
LANL	Los Alamos National Laboratory
LLNL	Lawrence Livermore National Laboratory
NNSS	National Nuclear Security Site
PNNL	Pacific Northwest National Laboratory
RPT	Radiological Protection Technologist
TIMS	Thermal Ionization Mass Spectrometry
TRU	Trans-Uranic

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1.0 Introduction

In FY16, four Pu targets were fabricated in support of the Nuclear Physics: Cumulative Fission Product Yields (Fission R-values) task of the NA-22 sponsored F2016 venture project; these targets are suitable for irradiation in the Flattop critical assembly at the Nevada National Security Site (NNSS). Two of the targets contain 99% ^{239}Pu while two targets contain 93% ^{239}Pu. These targets utilize the design developed in FY12 under the Nuclear Physics: Cumulative Fission Product Yields (Fission R-values) task of the NA-22 sponsored F2012 venture project. The target design uses a double enclosure to ensure no Pu is released from the target in which an inner aluminum cup holding the Pu metal which is sealed in an outer stainless-steel capsule. The targets fit in the sample holders used with the Flattop critical assembly and can be incorporated into a standard Flattop irradiation like those routinely performed on these projects. This report provides an overview of the target design, preparation of the Pu metal materials, target fabrication, target assay, and target leak testing.

2.0 Target Design

In FY12, under the F2012 project, a target design was developed at PNNL suitable for irradiation of Pu on the Flattop critical assembly at the NNSS. To ensure adequate containment meeting the requirements of the NNSS facility, the target consists of the Pu metal held in an aluminum cup which is sealed within a stainless-steel capsule providing double containment of the Pu metal.

The aluminum cup consists of two pieces, a short bottom cup, and a longer top cup as shown in Figure 1 parts a and b. A press was used to close the aluminum cup by folding the longer cup over the shorter cup, as described in section 4.1; a sealed aluminum cup is shown in Figure 1 part c. The aluminum cup provides a cover for the pyrophoric Pu metal and initial containment to prevent the spread of radioactive material while sealing the stainless-steel capsule. It is important to note the aluminum cup is very thin and easily bent or punctured so it must be handled with care. A drawing of Pu metal pieces contained in the cup as well as a sealed cup containing Pu are shown in Figure 1 part d.

The stainless-steel capsules, shown in Figure 1 parts e through h, were machined to fit the Al cups described above and sealed by welding an outer lip. The capsule can be opened by cutting along the welded edge without contacting the aluminum wrapped Pu metal.

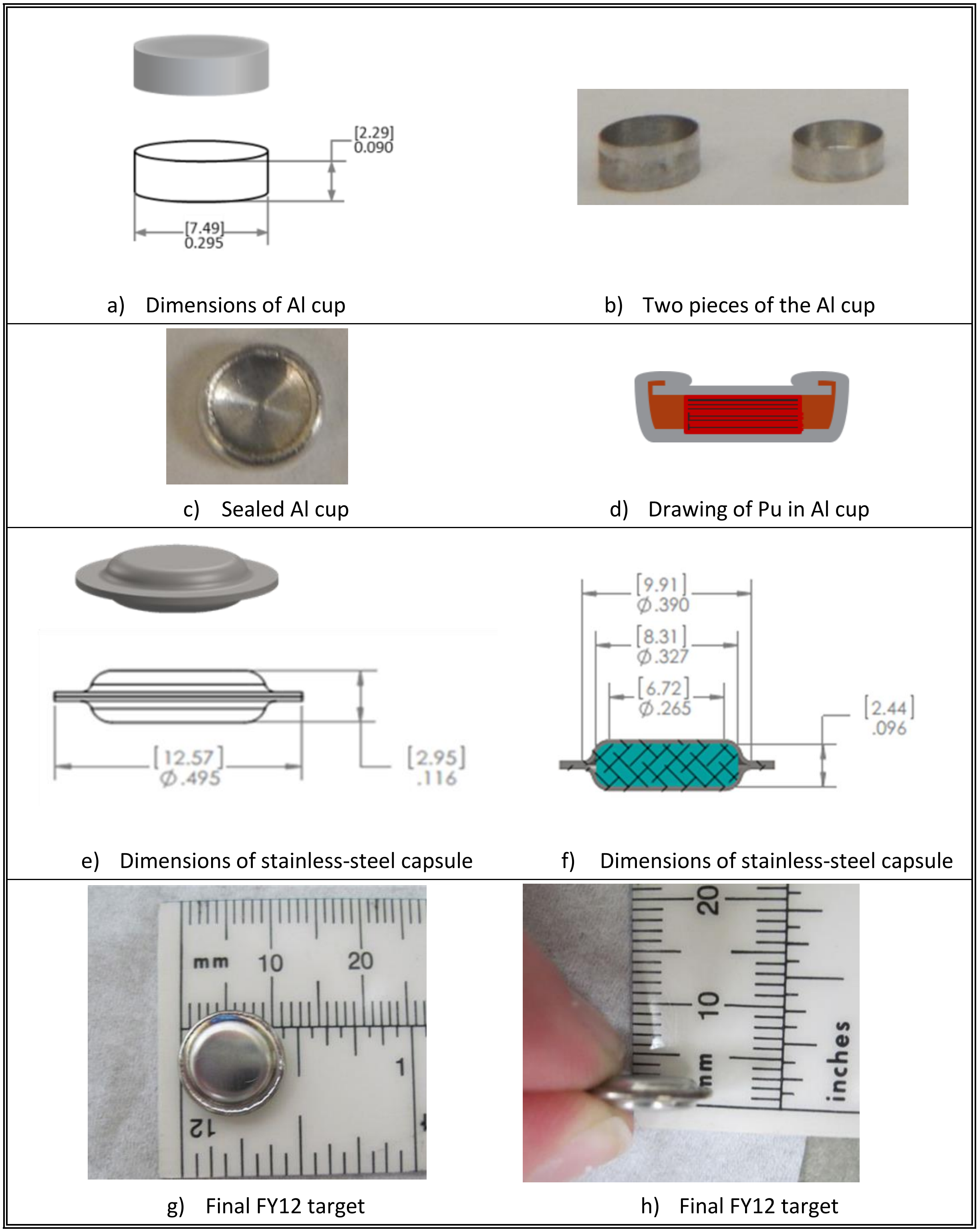


Figure 1. Components of the Pu target including the Al cup and stainless-steel capsule

3.0 Preparation of Plutonium Materials

3.1 Overview

Four targets were fabricated with two containing 99% ^{239}Pu metal and two containing 93% ^{239}Pu metal. The 99% ^{239}Pu metal was in the form of foils while the 93% ^{239}Pu was in larger chunks. Both materials were coated with an oxide layer which was removed before weighing the material used in each target to obtain an accurate Pu mass for each target. Additional metal pieces were cleaned, weighed, and stored in separate containers from the targets. These additional samples provided clean samples of each material for additional analyzes such as thermal ionization mass spectrometry (TIMS).

The cleaning process used a Dremel tool to physically remove the oxide layer from the surface of the Pu metal. The use of a Dremel generated significant quantities of ultrafine Pu metal dust which is pyrophoric. To prevent reaction of the pyrophoric Pu dust with atmospheric oxygen and to contain the significant amount of loose radioactive material, the cleaning process was accomplished using disposable two chambered glove bags, purchased from LANCS, located in a radiologically controlled contamination area (CA) fume hood. The glove bags were purged with argon and an oxygen sensor was used to ensure minimal oxygen presence in the glove bag. To ensure sufficient quantities of argon were available during the cleaning process, a manifold was setup in the laboratory with multiple argon cylinders allowing for easy switching between cylinders.

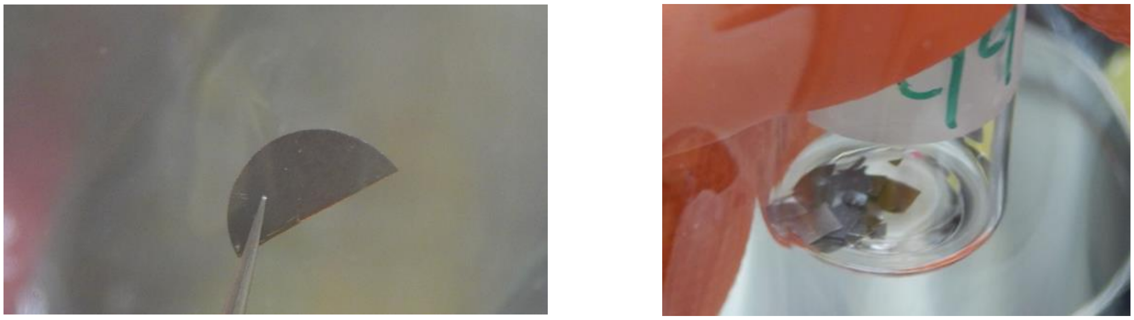
Following the cleaning process for the 99% ^{239}Pu metal foils, the foils were cut into small strips for loading into the aluminum cups. After completing the preparation of the 99% ^{239}Pu metal, the 93% ^{239}Pu metal was cleaned. The Dremel was then used to shape the larger chunks into pieces that would fit into the aluminum cups. The clean Pu metal was maintained under argon by sealing the containers with tape or dental wax. The vials containing the Pu metal pieces were wiped clean and removed from the glove bag.

After the cleaning and shaping process was complete, the glove bag contained around 2.5 grams of ultrafine Pu dust. To prevent the pyrophoric dust from igniting, the argon flow to the glove bag was shut off and the bag was allowed to sit over the weekend to allow for slow oxidation of the Pu dust. After roughly 2.5 days, the oxygen level in the glove bag was at atmospheric quantities and the Pu dust was considered to be oxidized and no longer pyrophoric. The glove bag was then placed in a waste drum and a new glove bag was setup for loading the Pu metal into the aluminum cups.

3.2 Pu-239 metal – 99%

The 99% ^{239}Pu metal, shown in Figure 2, was a thin 0.5 inch diameter foil with a dark oxide layer; this material was identified in the inventory system as 9P22d batch 453B. The material isotopics and results of the gamma energy analysis (GEA) assay are provided in section 5.2.

The oxide layer was removed by grinding with a Dremel 4000 series corded rotary tool, using a ¼ inch aluminum oxide pointed cone shaped grinding stone; portions of the Pu foil pre- and post-removal of the oxide layer are shown in Figure 2. Once the black oxide layer was removed, the metal was cut into pieces that were small enough to fit into the aluminum cup.



a) Pu metal prior to cleaning

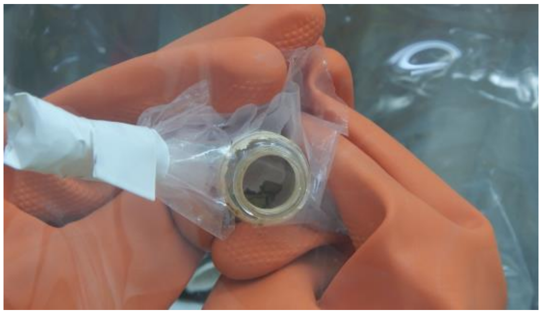
b) Pu metal post cleaning and cutting

Note: Photos were taken through the plastic glove bag which can result in distortion of the shape and color

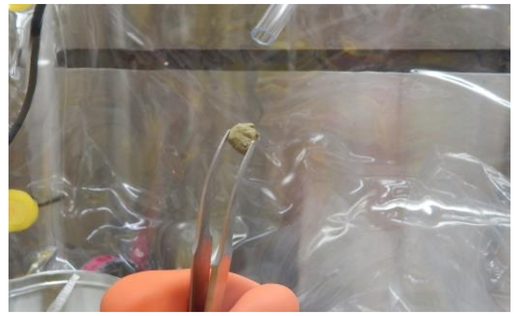
Figure 2. 99% ^{239}Pu metal before and after removal of oxide layer

3.3 Pu-239 metal – 93%

The stock 93% ^{239}Pu metal, obtained from Lawrence Livermore National Laboratory (LLNL) in 2006, was a chunky material covered in a green oxide layer, shown in Figure 3 parts a and b. The oxide layer was removed using a Dremel 4000 series corded rotary tool, using a ¼ inch aluminum oxide pointed cone shaped grinding stone, shown in Figure 3 parts c and d. The metal piece was held with a locking pair of pliers and ground until the oxide layer was completely removed, leaving a shiny metallic surface. The sample was further ground to a rough sphere, removing any sharp edges that could potentially perforate the aluminum cups. The final Pu metal pieces used in targets 3 and 4 are shown in Figure 3 parts e and f. The material isotopics and results of the GEA assay are provided in section 5.3.



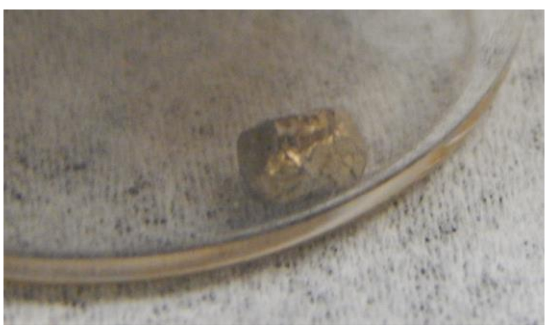
i) Stock 93% ^{239}Pu metal



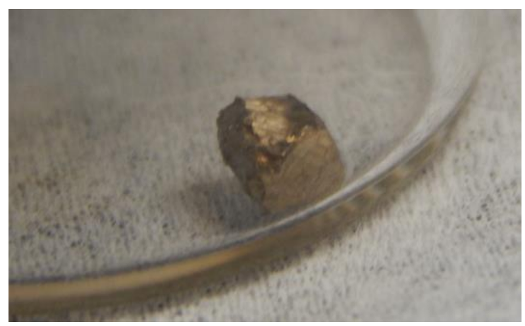
j) A piece of the 93% ^{239}Pu metal prior to removal of the oxide layer

k) Use of a Dremel to remove the oxide layer from the Pu metal

l) A partially cleaned piece of the 93% ^{239}Pu metal



m) Cleaned 93% Pu-239 metal used for target #3



n) Cleaned 93% Pu-239 metal used for target #4

Note: Photos were taken through the plastic glove bag which can result in distortion of the shape and color

Figure 3. 93% Pu-239 metal with oxide layer, the cleaning process, and the final metal used in targets #3 and #4

4.0 Fabrication of Plutonium Targets

4.1 Loading Pu metal into Al cups

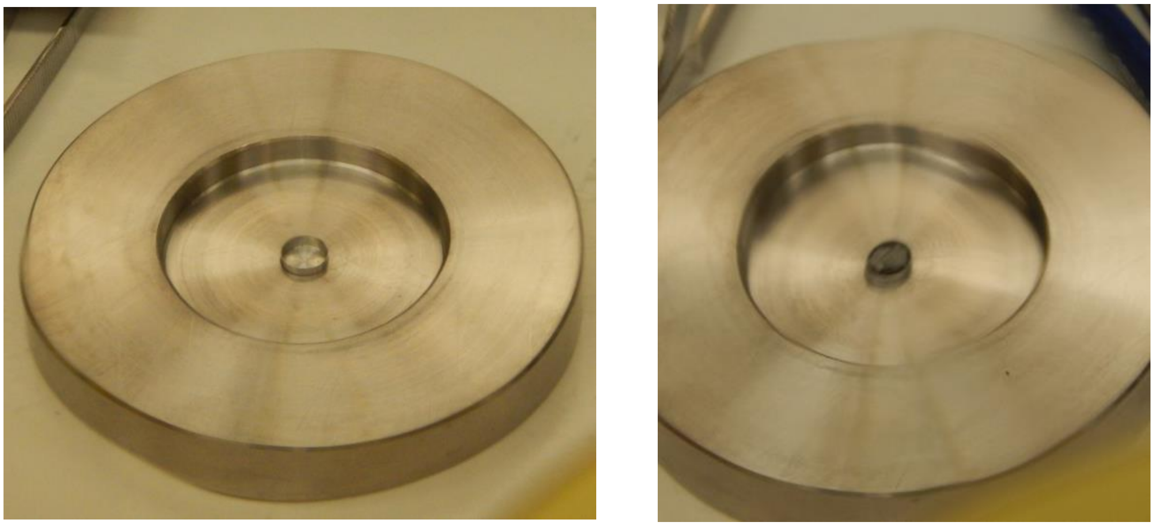
After cleaning the Pu material, a new glove bag was set up for loading the Pu metal into the aluminum cups to keep radiological contamination present in the glove bag to a minimum; the loaded aluminum cups had to be free of any external radiological contamination prior to welding the outer stainless steel capsule in a non-radiological fume hood. The use of a new glove bag also prevented cross contamination between the two Pu materials since the cleaning process results in significant quantities of Pu metal dust. The 99% ^{239}Pu targets were prepared first followed by the 93% ^{239}Pu targets. A 4-place calibrated analytical balance was used to ensure an appropriate amount of Pu metal was in the cup.

The loading process was started by obtaining the mass of the top and bottom of each aluminum cup so that an accurate mass for the Pu metal could be obtained by weighing the cup post loading. The aluminum cups were machined at PNNL from high purity aluminum.

The necessary equipment was setup in the new glove bag including the loading fixture and press. The loading fixture was used for placing the Pu metal into the bottom aluminum cup without introducing radiological contamination to the exterior of the cup and the press was used for closing the aluminum cup by crimping the edge of the top aluminum cup.

The bottom aluminum cup was centered in the loading fixture, as shown in Figure 4. Once the cup was in the loading fixture, the top portion of the loading fixture was placed over the cup; this portion of the loading fixture had a funnel which sat over the aluminum cup so that the Pu material was funneled into the cup and did not touch the top or outer portion of the cup.**Error! Reference source not found.** Pieces of the 99% ^{239}Pu metal were carefully dropped through the funnel until the cup was packed as full as possible; an aluminum cup containing 99% ^{239}Pu is shown in Figure 4.

After carefully removing the funnel, the top aluminum cup was placed over the bottom aluminum cup. The top cup was longer than the bottom cup so that it hung past the edge of the bottom cup; this edge is what was crimped to seal the cup. There was very little difference in the diameters of the top and bottom cups so it was necessary that the top cup be placed onto the bottom cup completely straight then tapped down until the cup was as closed as possible while sitting in the loading fixture. Once the top of the cup had been pushed down as far as possible, the cup was carefully picked up using tweezers; the aluminum cups are very thin and easily bent so it is critical that too much pressure is not applied when handling the cups. A pencil was used to push the bottom portion of the cup up completely into the top portion of the cup.



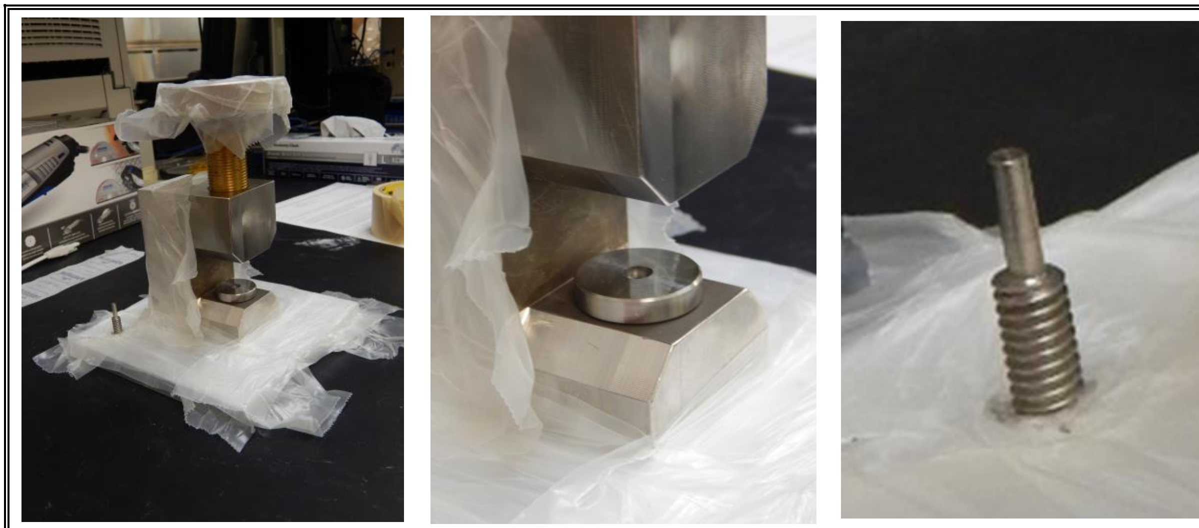
a) Cleaned 93% Pu-239 metal used for target #3

b) 99% ^{239}Pu pieces loaded into bottom aluminum cup inside loading fixture

Note: Photos were taken through the plastic glove bag which can result in distortion of the shape and color

Figure 4. Fixture for loading Pu metal into the bottom Al cup

The cup was sealed using the screw press shown in Figure 5**Error! Reference source not found.**; the screw presses down on the cup sealing it by rolling the ends of the longer top cup over and into the bottom of the short cup. The body of the press was made of aluminum while the screw was made of brass. After removing the cup from the loading fixture, it was turned upside down then loaded into the screw press where it sat inside an aluminum holder. The brass screw was then turned until it could not turn anymore. The holder containing the cup was carefully removed and placed over the extractor that was built into the base of the press. The holder was screwed onto the extractor until the cup popped out a bit at which point it was removed with tweezers. The sealed Al cups for targets #1 and #2 are shown in Figure 6.



a) Screw press

b) Close-up of Al holder

c) Extractor

Figure 5. The target screw press with the aluminum holder and extractor

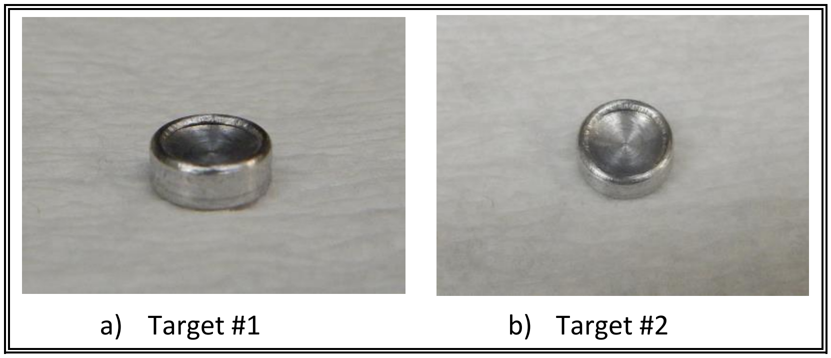


Figure 6. Sealed aluminum cup containing Pu for target #1 and target #2

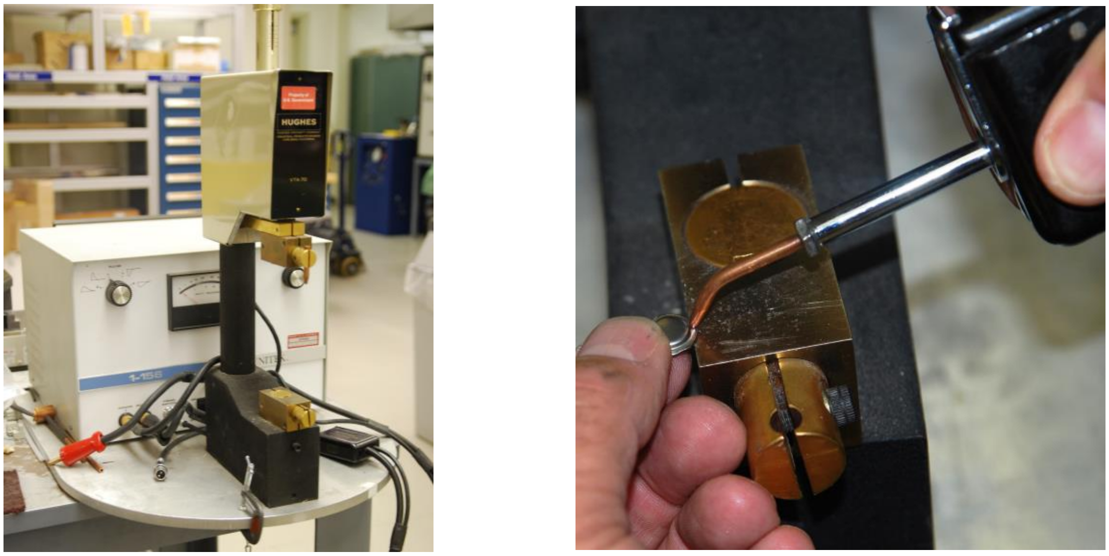
Each sealed cup was carefully smeared for radiological contamination per the RPTs instructions. The sealed cup was carefully smeared and placed onto a clean towel in the glove bag port. Once it was determined that the cup was free of external contamination, it was removed from the glove bag port and placed on a clean towel inside the fume hood; if contamination was found the cup was carefully wiped down and re-smeared. A direct survey was then performed on the top and bottom of the cup. For cups with persistent contamination after cleaning, a piece of very thin high purity aluminum foil was used to contain the contamination and the cup surveyed again; if needed the cup was torn open with tweezers to remove the Pu metal which was then loaded into a new cup. Once the sample was confirmed to be completely free of contamination, it was placed inside a plastic jar and allowed to sit in the fume hood until it was ready to be placed into the stainless-steel outer containers which were then welded. Each cup was removed just prior to welding.

After preparing the aluminum cups with 99% ^{239}Pu, the two 93% ^{239}Pu cups were prepared. The 93% material was formed into roughly spherical pieces of Pu metal during the cleaning process making loading them into the cups easier than the small strips used for the 99% material. Because of their size and spherical nature, the aluminum cups slightly bulged after sealing resulting in micro-holes in the aluminum, invisible to the eye, but enough to measure alpha radiation with an instrument held over the samples. Although the samples were found to be radiologically clean when smeared, alpha radiation was being released through the micro-holes. In order to ensure that no radiation, or eventually, contamination, would be able to penetrate the aluminum, the samples were further wrapped in a layer of 99.999% aluminum foil. The samples were then determined to be free of external contamination as well as no longer having any release of alpha radiation. These two samples were placed inside plastic jars and left in the fume hood until they were removed to be placed into the stainless-steel outer containers and welded.

4.2 Target welding

A spot welder and orbital welder were used to seal the stainless-steel outer containers; the stainless-steel containers were machined at PNNL. These were located in a non-radiological fume hood to prevent the equipment from requiring radiological control. This work was performed with continuous RPT coverage with the RPT surveying the target at each step in the process to ensure the target had not been breached. In the event that a target was found to have alpha contamination, the fume hood would have been up-posted to a CA and work halted.

A spot welder, Hughes Model HRW-750B, was used to weld 6 to 8 points around the outside of the stainless-steel capsule in order to hold the capsule close during the welding process with the orbital welder. A picture of the spot welder with power supply and spot-welding a non-radiological target are shown in Figure 7.

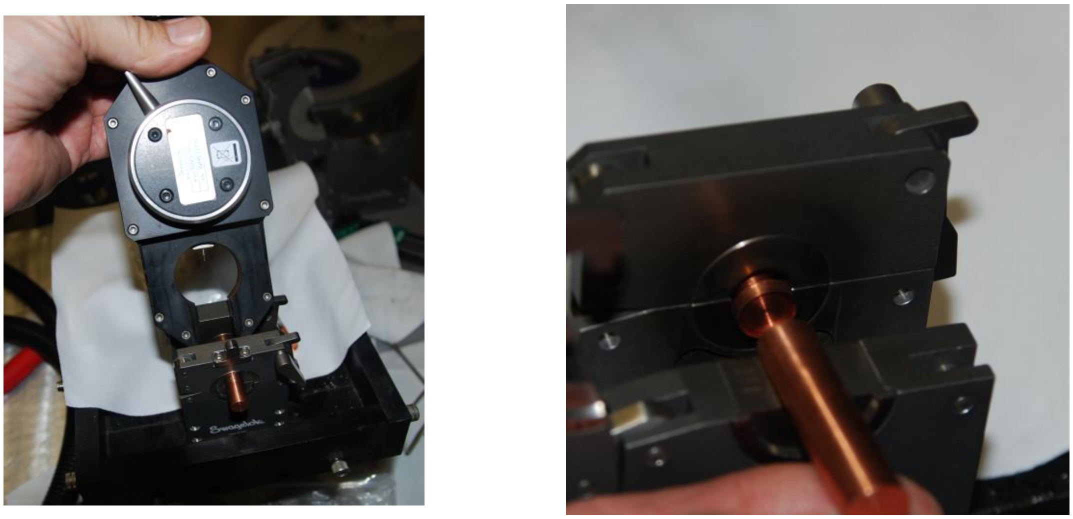


a) Spot welder with power supply

b) Demonstration of location for spot welds using a simulated non-radiological target

Figure 7. Spot welder

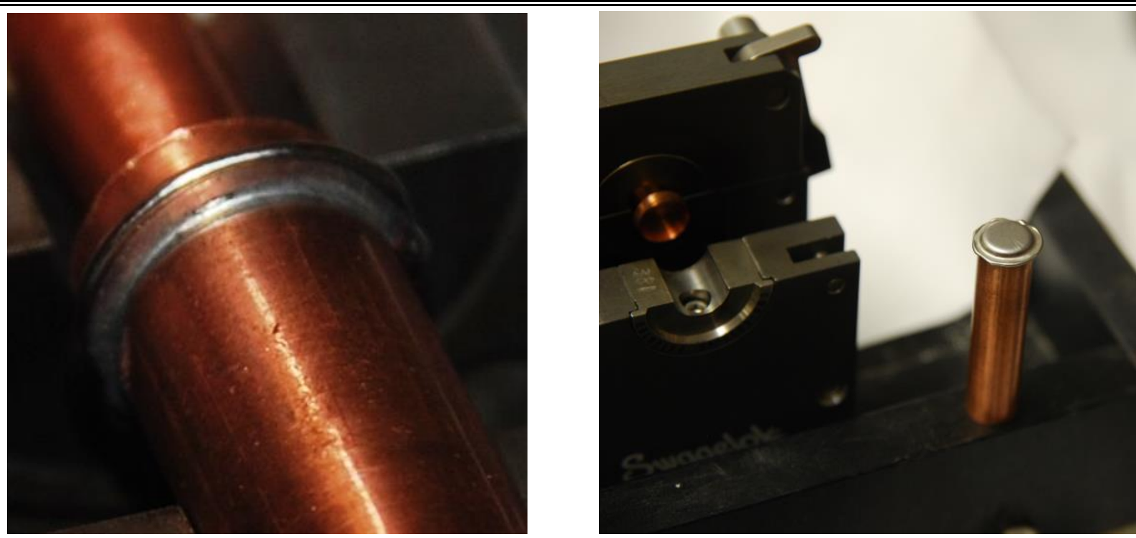
The stainless-steel capsules were sealed using a Swagelok M200 Welding System orbital welder. The target was held in place with a copper fixture, which also acted as a heat sink. A picture of the orbital welder and the copper fixture are shown in Figure 8 and the stainless-steel capsule held in the copper fixture and the target post welding are shown in Figure 9. Initially a lower power level was used during the welding such that the heat input was high enough to fuse the edges of the capsule halves after which the power level was increased to melt the edges of the capsule in order to form a nice weld bead. The parameters used on the orbital welder are given in Table 4-1.



a) Orbital welder with copper fixture

b) Close up of copper fixture

Figure 8. (a) Orbital welder; (b) Close up of copper fixture



a) Stainless-steel target in the copper weld fixture

b) Stainless-steel target on the copper weld fixture after sealing

Figure 9. Stainless-steel target with the copper weld fixture

Table 4-1. Parameters for Orbital Welder

Parameter	Weld 1	Weld 2
High Power, Amps	6.0	8.0
Low Power, Amps	5.5	5.5
Weld Time, Seconds	12.5	12.5
Ramp Time, Seconds	0.5	0.5
Pulse Rate, Hz	10.0	10.0
Pulse Width, Percent	50	50
Hi Power Speed, rpm	5.00	5.00

Parameter	Weld 1	Weld 2
Low Power Speed, rpm	5.00	5.00
Average Power, Amps	5.8	6.8
Purge gas	Argon	Argon
Purge rate, cfm	20	20
Electrode Material	Tungsten	Tungsten

4.3 Final targets

The final welded targets are shown in Figure 10. In order to accommodate this increased size of the 93% plutonium targets, the stainless-steel cups were slightly enlarged by hammering out the raised portion of them.

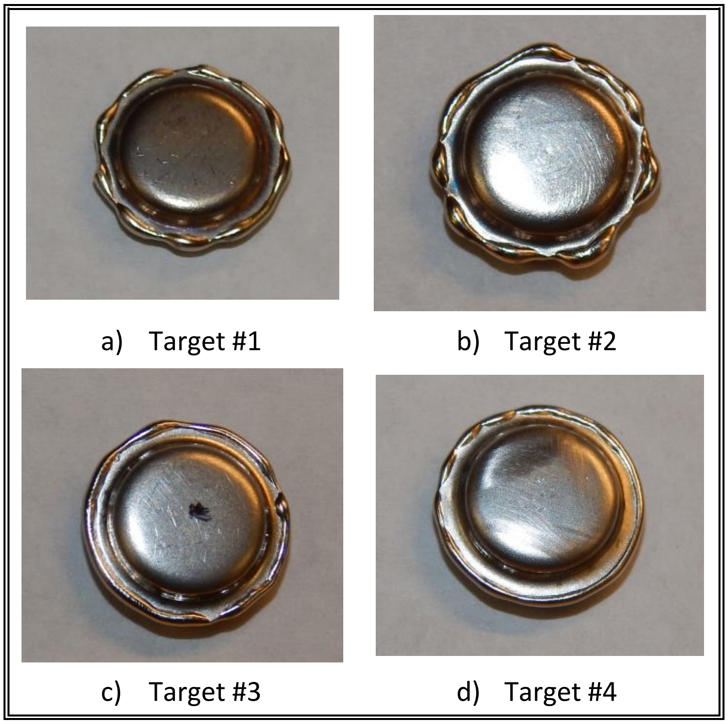


Figure 10. Final welded targets for targets 1, 3, and 4 with the first iteration of target 2; visual inspection of target 2 failed and it was opened and sealed in a new stainless-steel capsule; targets 1 and 2 contain 99% ^{239}Pu while targets 3 and 4 contain 93% ^{239}Pu

4.4 Helium leak check

To ensure the targets were tightly sealed and would not leak Pu, the four targets were tested using a helium leak test. The leak test was performed using a Pfeiffer HLT550 helium leak detector and a calibrated helium leak supplied by Vacuum Technology Incorporated. Prior to testing, the targets were placed in a helium bombing chamber. The chamber was pumped down to a pressure of 2x10^{-7} Torr, and then pressurized to 1635 Torr with helium. The targets were exposed to the helium atmosphere for 24 hours. At the end of this time, the helium pressure was 1623 Torr. The targets were then removed from the chamber and allowed to de-gas for one hour before the helium leak rate analysis was performed. Leak rate analysis was performed by taking alternating measurements of the instrument background, the calibrated leak, and the targets. The targets were found to have average normalized helium leak rates of less than 1.0x10^{-9} atm cc/sec. The leak test report is shown in section 8.0Appendix A.

5.0 Plutonium Target Material and Assay

5.1 Final target and sample masses

Two targets were prepared with 99% ^{239}Pu metal and a small amount of the same material was set aside for TIMS analysis at PNNL to verify the historical isotopic assay, the final masses are given in

Table 5-1. The three samples were each assayed by GEA with the isotopic results shown in Table 5-5 and Table 5-6. The sample set aside for TIMS has been dissolved but has not been analyzed yet.

Table 5-1. Final Pu masses for 99% Pu-239 samples

Sample ID	RMT ID	Pu Mass, g		
		Weighed	GEA	
67655-22-Pu1	90045	0.2475	0.258	Target
67655-23-Pu2	89994	0.3313	0.323	Target
67655-28-Pu7	90124	0.0125	0.0411	PNNL TIMS
67655-105-5	94682	0.0310	N/A	LANL MS

Two targets were prepared with 93% ^{239}Pu metal and two samples of the same material were set aside for TIMS analysis at PNNL and LANL to verify the historical isotopic assay, the final masses are given in Table 5-2. The four samples were each assayed by GEA with the isotopic results shown in Table 5-8 and Table 5-9. The sample set aside for TIMS analysis at PNNL has been dissolved but has not been analyzed as of FY20 while the sample set aside for TIMS analysis at LANL has not been dissolved.

Table 5-2. Final Pu masses for 93% Pu-239 samples

Sample ID	RMT ID	Pu Mass, g		
		Weighed	GEA	
67655-24-Pu3	90064	0.2301	0.255	Target
67655-25-Pu4	90065	0.1979	0.186	Target
67655-26-Pu5	90066	0.3124	0.188	PNNL TIMS
67655-27-Pu6	90067	0.3605	0.243	LANL MS

Since the oxide layer was removed prior to weighing, the weight determined using a calibrated analytical balance should be used as the final mass. When comparing the final sample masses determined by weight and by GEA assay, it should be noted that the samples are not in calibrated geometries and therefore self-absorption of the gamma rays due to the sample thickness was an issue. The 93% ^{239}Pu samples were thicker than the 99% ^{239}Pu samples due to the small round ball shape rather than metal foils, therefore there is more variation in the measured weight compared to the GEA results for the 93% material. There is also a significant difference in sample 7, the 99% ^{239}Pu for PNNL TIMS analysis, which is primarily due to the small amount of material measured in an uncalibrated geometry.

The GEA results were corrected for gamma self-absorption using a semi-empirical approach to obtain the best fit to gamma emissions as a function of gamma energy from ^{239}Pu. The self-absorption corrections were very large for the lower energy gamma from ^{241}Am. The activities were then converted to mass and Pu wt% using the specific activities given in Table 5-3.

Table 5-3. Specific activities for Pu and Am isotopes

Isotope	Specific Activity (Bq/g)
^{238}Pu	6.334×10^{11}
^{239}Pu	2.295×10^{9}
^{240}Pu	8.398×10^{9}
^{241}Pu	3.811×10^{12}
^{242}Pu	1.465×10^{8}
^{241}Am	1.268×10^{11}

Two pieces of the 93% Pu metal were set aside for TIMS analysis at PNNL and LANL, 67655-26-Pu5 and 67655-27-Pu6, respectively. On May 12th 2017, the Pu piece set aside for PNNL was dissolved in 50% HNO_3 – 50% HCl and split among multiple sample bottles; the final solution and Pu masses are given in

Table 5-4. Two samples were designated to go for TIMS analysis in 3430; one of the samples was supposed to be a blank from the dissolution process but there was a miscommunication and both aliquots contain the Pu solution.

Table 5-4. Final solution and Pu masses for splits of 67655-27-Pu7

Sample ID	RMT ID	Solution Mass (g)	Pu Mass (mg)	
Pu-239 KLN-2	92806	10.5757	25.6857	PNNL TIMS
Pu-239 KLN-1	92807	10.6380	25.8371	PNNL TIMS
67655-26-Pu5A	92805	10.7597	26.1326	
67655-26-Pu5B	92804	10.6924	25.9692	
67655-26-Pu5C	92803	10.7007	25.9893	
67655-26-Pu5D	92802	10.7501	26.1093	
67655-26-Pu5E	92801	10.7113	26.0151	
67655-26-Pu5F	92800	10.7120	26.0168	
67655-26-Pu5G	92799	10.7299	26.0602	
67655-26-Pu5H	92798	10.7530	26.1163	
67655-26-Pu5I	92797	10.7192	26.0342	
67655-26-Pu5J	92796	10.5417	25.6031	

5.2 Pu-239 – 99%

The 99% ^{239}Pu metal, shown in Figure 2, was a thin 0.5 inch diameter foil with a dark oxide layer; this material was identified as 9P22d batch 453B. The results from the GEA assay in FY16 are shown in Table 5-5 while Table 5-6 provides the weight percent for the various Pu isotopes based on the historical assay and the FY16 GEA assay. The complete historical assay information is given in Table 8-1.

It should be noted that the GEA assay of 67655-22-Pu1 suggests the target material is 93% ^{239}Pu rather than 99%. The results of this assay are extremely close to the results for 67655-24-Pu3 which is 93% ^{239}Pu so there may have been a mix-up with the samples. The target material was not mixed up since the 99% ^{239}Pu material were small foil pieces while the 93% ^{239}Pu material had to be shaped into small balls from a larger chunk. If desired, the target could be reanalyzed prior to irradiation.

Table 5-5. Results for the GEA assay of the 99% Pu target material

Sample ID	67655-22-Pu1		67655-23-Pu2		67655-28-Pu7	
Assay Date	10/5/2016		11/16/2016		10/5/2016	
	Bq/sample ± 1σ%	Wt (g)	Bq/sample ± 1σ%	Wt (g)	Bq/sample ± 1σ%	Wt (g)
^{239}Pu	$5.51x10^{8}$ ± 2%	$2.40x10^{-1}$	$7.31x10^{8}$ ± 2%	$3.18x10^{-1}$	$9.29x10^{7}$ ± 2%	$4.05x10^{-2}$
^{240}Pu	$1.52x10^{8}$ ± 2%	$1.81x10^{-2}$	$4.19x10^{7}$ ± 6%	$4.99x10^{-3}$	$5.65x10^{6}$ ± 27%	$6.73x10^{-4}$
^{241}Pu			$4.30x10^{6}$ ± 29%	$1.13x10^{-6}$		
^{241}Am	$3.23x10^{6}$ ± 2%	$2.55x10^{-5}$	$4.51x10^{6}$ ± 4%	$3.56x10^{-5}$	$6.07x10^{5}$ ± 2%	$4.11x10^{-2}$
Total Pu		0.258		0.323		0.0411

Table 5-6. Weight percent for the various Pu isotopes in the 99% Pu target material (Weight % ± 1σ%)

Sample ID	Historical Assay		67655-22-Pu1	67655-23-Pu2	67655-28-Pu7
Assay Date	5/23/1972	Decay Corrected to 10/5/2016	10/5/2016	11/16/2016	10/5/2016
^{238}Pu	< 0.002				
^{239}Pu	99.11 ± 0.01	99.11	93.0	98.5	98.4
^{240}Pu	0.880 ± 0.005	0.88	7.01	1.54	1.64
^{241}Pu	0.010 ± 0.001	0.001181		$3.50x10^{-4}$	
^{242}Pu	0.005 ± 0.001	0.005			
^{244}Pu	< 0.0005				
^{241}Am		0.00882			

5.3 Pu-239 – 93%

The 93% 239Pu material, shown in Figure 3, was a chunky material with an oxide layer. This material was provided to PNNL by LLNL in 2006. The elemental composition of the Pu material based on historical documentation provided by LLNL is given in Table 5-7. The results from the GEA assay in FY16 are shown in Table 5-8 while Table 5-9 provides the weight percent for the various Pu isotopes based on the historical assay and the FY16 GEA assay. The complete historical assay information is given in Table 8-2.

It should be noted that the material has a large quantity of gallium and a fair amount of iron. The presence of activation products of gallium and iron should be considered when performing radiometric analysis of the irradiated sample since activation products of these elements are not typically seen in actinide targets irradiated on the Flattop critical assembly. The gallium and iron could also impact the chemical separations performed prior to radiometric analyses.

Table 5-7. Elemental composition of 93% Pu target material given in the historical documentation

Element or Isotope	Reference Alloy #2	Minimum Detection Limit	Element or Isotope	Reference Alloy #2	Minimum Detection Limit
Al	55(2) µg/g	20	^{232}Th	Not detected	0.2
V	Not detected	2	^{233}U	Not detected	0.2
Cr	2.8(3) µg/g	1	^{234}U	1.3(9) µg/g	0.2
Mn	3.8(3) µg/g	1	^{235}U	14(3) µg/g	0.2
Fe	293(8) µg/g	5	^{237}Np	70(1) µg/g	0.2
Ni	4(1) µg/g	1	^{238}Pu, ^{238}U	0.011(3) wt%	0.001
Cu	2(1) µg/g	1	^{239}Pu	92.93(3) wt%	0.05
Ga	9727(99) µg/g	5	^{240}Pu	5.73(3) wt%	0.001
Y	14(5) µg/g	0.2	^{241}Pu, ^{241}Am	0.263(1) wt%	0.0001
Ta	37(13) µg/g	0.2	^{242}Pu	0.0304(2) wt%	0.00005
W	119(41) µg/g	0.2			

Table 5-8. Results for the GEA assay of the 93% Pu target material

Sample ID	67655-24-Pu3		67655-25-Pu4		67655-26-Pu5		67655-27-Pu6	
Assay Date	10/5/2016		10/5/2016		10/5/2016		10/5/2016	
	Bq/sample ± 1σ%	Wt (g)	Bq/sample ± 1σ%	Wt (g)	Bq/sample ± 1σ%	Wt (g)	Bq/sample ± 1σ%	Wt (g)
^{239}Pu	$5.46x10^{8}$ ± 2%	$2.38x10^{-1}$	$3.97x10^{8}$ ± 2%	$1.73x10^{-1}$	$4.03x10^{8}$ ± 2%	$1.76x10^{-1}$	$5.21x10^{8}$ ± 2%	$2.27x10^{-1}$
^{240}Pu	$1.41x10^{8}$ ± 7%	$1.58x10^{-2}$	$1.06x10^{8}$ ± 6%	$1.26x10^{-2}$	$1.06x10^{8}$ ± 14%	$1.26x10^{-2}$	$1.33x10^{8}$ ± 18%	$1.59x10^{-2}$
^{241}Pu	$6.57x10^{8}$ ± 4%	$1.72x10^{-4}$	$5.01x10^{8}$ ± 3%	$1.32x10^{-4}$	$5.04x10^{8}$ ± 4%	$1.32x10^{-4}$	$6.56x10^{8}$ ± 9%	$1.72x10^{-4}$
^{241}Am	$4.64x10^{7}$ ± 12%	$3.66x10^{-4}$	$3.97x10^{7}$ ± 2%	$3.13x10^{-4}$	$4.05x10^{7}$ ± 10%	$3.19x10^{-4}$	$5.22x10^{7}$ ± 2%	$4.11x10^{-4}$
Total Pu		0.255		0.186		0.188		0.243

Table 5-9. Weight percent for the various Pu isotopes in the 93% Pu target material (Weight % ± 1σ%)

	Historic	67655-24-Pu3	67655-25-Pu4	67655-26-Pu5	67655-27-Pu6
Assay Date		10/5/2016	10/5/2016	10/5/2016	10/5/2016
^{238}Pu	0.0113				
^{239}Pu	92.93	93.3	93.1	93.2	93.4
^{240}Pu	5.73	6.60	6.78	6.71	6.53
^{241}Pu	0.2631*	0.0677	0.0708	0.0702	0.0709
^{242}Pu	0.005				

*Value provided in the historical documentation includes both ^{241}Pu and ^{241}Am

6.0 Documentation

6.1 Travelers

A traveler or traveler paper is required to be located with each target. This paper documents who prepared each target, inspected the target, and released the target from radiological control. The travelers for all the targets were scanned and electronic copies were loaded onto the project share drive for project records. The documents can also be seen in section 8.0Appendix C.

6.2 Off-site shipment

If shipping the targets off-site, they must be surveyed for exterior contamination prior to packaging. The target(s) should be shipped with the He leak test information and traveler(s). The He leak test results and any other requested documentation should be sent to the irradiation facility prior to any shipment.

6.3 Procedures and permits

The following procedures and safety documentation were utilized during production of the Pu targets:

- Radiological Work Permit NSD-508 Rev 03
- RPL-GB-008 Rev 04, Preparation of Pyrophoric Metals Under Inert Conditions
- RPL-WELD-01 Rev 01, NSD Welding Small Targets for Irradiation Experiments

7.0 Lessons Learned and Recommendations

7.1 Aluminum cups and foil

- The aluminum cups were machined at PNNL from high purity aluminum. The aluminum cups must be high purity to avoid forming unwanted activation products.

- Aluminum foil may be needed if a small leak is discovered after the sample has been pressed into the aluminum cup. The aluminum foil must be high purity to avoid forming unwanted activation products.

- The aluminum cups are very thin and easily bent so it is critical that too much pressure is not applied when handling the cups. A pencil was used to push the bottom of the cup up into the top all the way. The cup was then carefully inverted and placed onto the press and sealed. The holder for the cup was removed from the press and placed onto the extractor and pushed out until it could be removed.

7.2 Glove bags and Pu cleaning process

- Cleaning the Pu material and loading the aluminum cups must be performed in separate glove bags due to the amount of loose Pu dust following the cleaning process using a Dremel. The glove bag used for cleaning the Pu metal should be double chambered while the glove bag used for loading the aluminum cups can be single chambered to accommodate the size of the press.

- All supplies should be set up in the glove bags prior to use, to include tape, a brush for cleaning inside the glove bag, and high purity aluminum foil if needed during loading of the aluminum cup.

- It would be useful to have a small, disposable balance available when cleaning and sizing the Pu material to get an approximate mass of Pu metal being loaded inside the aluminum cup. Otherwise, when the Pu is weighed it is already sealed in a pre-weighed aluminum cup; this involves moving the cup between CA fume hoods twice and removing it from the glove bag. If the weight is incorrect, the cup must be opened and a new cup prepared with the correct amount of Pu.

7.3 Waste

- The Pu metal cleaning process generated accountable quantities of trans-uranium (TRU) waste.

    - Staff should work with the field services representative (FSR) prior to generating TRU waste. There are additional requirements for generating TRU waste which impact the size and weight of what is removed from the CA fume hood.

    - Handling accountable amounts of material requires careful tracking of what mass of material is in each container so that the amount present in the glove bag sent for waste is accurate.

- Staff should work with the fissionable material supervisor to ensure the correct information is recorded and the radioactive material tracking system is appropriately updated for sample splits.

7.4 Post-irradiation processing

- The aluminum cup and foil (if used) should be dissolved with the Pu metal to ensure no recoil fission products are lost; using low purity aluminum will introduce activation products which could adversely impact the radiometric analysis of the samples. In FY12, kitchen foil was used during the target preparation so that in FY15 when this target was irradiated the aluminum was not dissolved with the target to avoid introducing a variety of activation products. Not dissolving the foil resulted in fractionation of the fission products due to loss of the recoil fission products embedded in the aluminum.

8.0 Conclusion

During FY16, 4 Pu targets suitable for irradiation in the Flattop critical assembly at the NNSS were fabricated at PNNL. Two targets contain 99% ^{239}Pu while two contain 93% ^{239}Pu. The Pu material was cleaned of all oxide and weighed prior to target fabrication. The targets were then assayed by GEA which was compared to historical values associated with each material. The targets were He leak tested to ensure the final welded seal was complete.

Appendix A – Helium Leak Test Report

MEMORANDUM

Pacific Northwest NATIONAL LABORATORY | Battelle The Business of Innovation

Date: April 4, 2017 Project No.:67655

To: Leah Arrigo

From: Crystal Rutherford Internal Distribution: File/LB

Subject: Helium leak testing of Pu targets

Leah,

The helium leak testing of 4 Pu targets has been completed. All 4 targets were found to have an average normalized helium leak rates value of less than 1.0 x10^{-9} atm cc/sec. The leak test was performed using a Pfeiffer HLT550 (WD58131) helium leak detector and a calibrated helium leak (6627) supplied by Vacuum Technology Incorporated. Helium leak testing was done following PNNL technical procedure RPL-HE-001, Rev. 0, *Dosimetry Capsule Helium Leak Testing*. A record of the analysis was recorded in LRB 61427 on page 68. Work package N64656 has been charged for the analysis.

Helium Soak

Targets were placed in a helium bombing chamber on April 3, 2017, and the chamber was pumped out to a system pressure of 2 x 10^{-7} Torr. Next, the chamber was pressurized to 1635 Torr with Helium. The targets were allowed to stand in the helium atmosphere for 24 hours, at the end of which, the helium pressure was 1623 Torr. Targets were then removed from the chamber and allowed to degas for 1 hour before the helium leak rate analysis was performed.

Leak Rate Analysis

Leak rate analysis was performed by taking alternating measurements of the instrument background, the calibrated leak and the targets. All targets were found to have an average normalized helium leak rate value of less than 1.0 x10^{-9} atm cc/sec.

If you have any questions, please contact me at 375-5576.

MEMORANDUM

Pacific Northwest NATIONAL LABORATORY | Battelle The Business of Innovation

Targets tested	Average Normalized Value (atm cc / sec)	Targets accepted	Targets rejected
All Targets (4 targets) 67655-22-Pu1 67655-23-Pu2 67655-24-Pu3 67655-25-Pu4	1.0 x10^{-9}	4 Targets	none

Concurrence: [signature] Date: 4/6/2017

E54-1900-001 (8/98)

Appendix B – Historical Sample Information for the Plutonium Metal used in FY16

Table 8-1. Historical information associated with the 99% Pu-239 material used to fabricate targets #1 and #2 in FY16

9P22d

(Batch 453B / Pu-239 Metal / 0.5 inch Diameter / Aluminum Wrapped)

Thickness	Foil No.	Pu Weight (mg)	1/28/2002 Total Ci	Pu-239 Ci	Pu-240 Ci	Pu-241 Ci	Pu-242 Ci	Am-241 Ci
5.7 mil	1	276.204	1.809E-2	1.698E-2	5.516E-4	4.790E-4	5.465E-8	7.880E-5
	3	262.825	1.722E-2	1.616E-2	5.249E-4	4.558E-4	5.201E-8	7.498E-5
	17	267.593	1.753E-2	1.646E-2	5.344E-4	4.641E-4	5.295E-8	7.634E-5
	18	262.710	1.721E-2	1.615E-2	5.247E-4	4.556E-4	5.198E-8	7.495E-5
	19	263.373	1.725E-2	1.620E-2	5.260E-4	4.568E-4	5.211E-8	7.514E-5
Total		**1332.705**	**8.731E-2**	**8.195E-2**	**2.662E-3**	**2.311E-3**	**2.637E-7**	**3.802E-4**

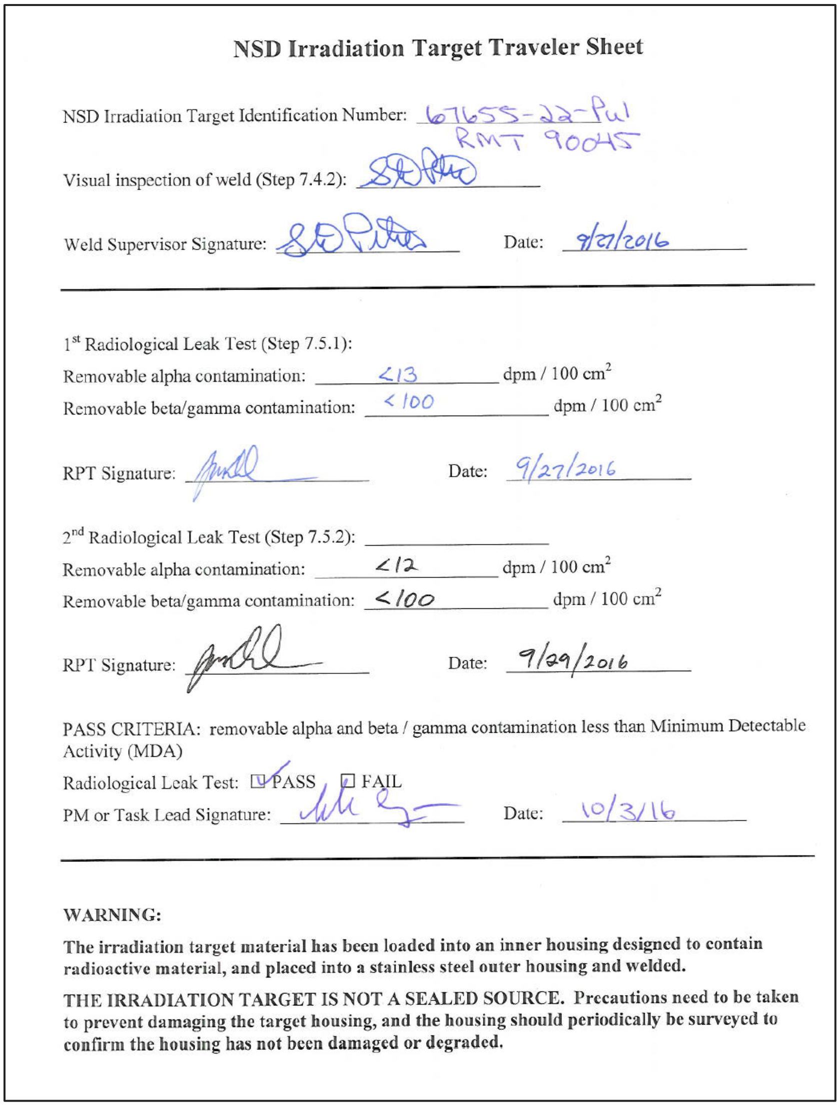
Note: Al can 0.021" thick; weight of Al about 200 mg/sample.

Isotope	5/23/1972 Pu Isotopes Wt%	+/-%	6/24/2009 Isotopes Wt%	SpA Ci/g	Ci/g of Sample
238	<0.002		<0.002	17.129	
239	99.11	0.01	99.11	0.062045	0.061493
240	0.880	0.005	0.88	0.22694	0.001997
241	0.010	0.001	0.001678	103.38	0.001734
242	0.005	0.001	0.005	0.0039575	1.979E-07
244	<0.0005				
Am-241	0		0.00832	3.428	0.00029

Table 8-2. Historical information associated with the 93% Pu-239 material used to fabricate targets #3 and #4 in FY16

Element or isotope	Reference alloy #2	Units	Minimum detection limit
Al	55(2)	wppm	20
V	ND	wppm	2
Cr	2.8(3)	wppm	1
Mn	3.8(3)	wppm	1
Fe	293(8)	wppm	5
Ni	4(1)	wppm	1
Cu	2(1)	wppm	1
Ga	9727(99)	wppm	5
Y	14(5)	wppm	0.2
Ta	37(13)	wppm	0.2
W	119(41)	wppm	0.2
^{232}Th	ND	wppm	0.2
^{233}U	ND	wppm	0.2
^{234}U	1.3(9)	wppm	0.2
^{235}U	14(3)	wppm	0.2
^{237}Np	70(1)	wppm	0.2
^{238}Pu,^{238}U	0.011(3)	wt %	0.001
^{239}Pu	92.93(3)	wt %	0.05
^{240}Pu	5.73(3)	wt %	0.001
^{241}Pu,^{241}Am	0.263(1)	wt %	0.0001
^{242}Pu	0.0304(2)	wt %	0.00005

Appendix C – Traveler Sheets



NSD Irradiation Target Traveler Sheet

NSD Irradiation Target Identification Number: 67655-22-Pu1 RMT 90045

Visual inspection of weld (Step 7.4.2): ____________

Weld Supervisor Signature: ____________ Date: 9/27/2016

1st Radiological Leak Test (Step 7.5.1):

Removable alpha contamination: <13 dpm / 100 cm^2

Removable beta/gamma contamination: <100 dpm / 100 cm^2

RPT Signature: ____________ Date: 9/27/2016

2nd Radiological Leak Test (Step 7.5.2): ____________

Removable alpha contamination: <12 dpm / 100 cm^2

Removable beta/gamma contamination: <100 dpm / 100 cm^2

RPT Signature: ____________ Date: 9/29/2016

PASS CRITERIA: removable alpha and beta / gamma contamination less than Minimum Detectable Activity (MDA)

Radiological Leak Test: ☑ PASS ☐ FAIL

PM or Task Lead Signature: ____________ Date: 10/3/16

WARNING:

The irradiation target material has been loaded into an inner housing designed to contain radioactive material, and placed into a stainless steel outer housing and welded.

THE IRRADIATION TARGET IS NOT A SEALED SOURCE. Precautions need to be taken to prevent damaging the target housing, and the housing should periodically be surveyed to confirm the housing has not been damaged or degraded.

Figure 11. Traveler sheet for target #1

NSD Irradiation Target Traveler Sheet

NSD Irradiation Target Identification Number: 67655-23-Pu2 RMT 899924

Visual inspection of weld (Step 7.4.2): ACCEPT

Weld Supervisor Signature: [signature] Date: 11/03/2016

1st Radiological Leak Test (Step 7.5.1):

Removable alpha contamination: < 20 dpm / 100 cm^2

Removable beta/gamma contamination: < 100 dpm / 100 cm^2

RPT Signature: [signature] Date: 11/03/16

2nd Radiological Leak Test (Step 7.5.2): —

Removable alpha contamination: <MDA – 15 dpm / 100 cm^2

Removable beta/gamma contamination: <MDA – 72 dpm / 100 cm^2

RPT Signature: T. Maves [signature] Date: 11/07/2016

PASS CRITERIA: removable alpha and beta / gamma contamination less than Minimum Detectable Activity (MDA)

Radiological Leak Test: ☑ PASS ☐ FAIL

PM or Task Lead Signature: [signature] Date: 11/7/2016

WARNING:

The irradiation target material has been loaded into an inner housing designed to contain radioactive material, and placed into a stainless steel outer housing and welded.

THE IRRADIATION TARGET IS NOT A SEALED SOURCE. Precautions need to be taken to prevent damaging the target housing, and the housing should periodically be surveyed to confirm the housing has not been damaged or degraded.

Figure 12. Traveler sheet for target #2

NSD Irradiation Target Traveler Sheet

NSD Irradiation Target Identification Number: 67655-24-Pu3 RMT 90064

Visual inspection of weld (Step 7.4.2): ________

Weld Supervisor Signature: ________ Date: 9/29/2016

1st Radiological Leak Test (Step 7.5.1):

Removable alpha contamination: <12 dpm / 100 cm^2

Removable beta/gamma contamination: <100 dpm / 100 cm^2

RPT Signature: ________ Date: 9/29/2016

2nd Radiological Leak Test (Step 7.5.2): ________

Removable alpha contamination: <13 dpm / 100 cm^2

Removable beta/gamma contamination: <1000 dpm / 100 cm^2

RPT Signature: ________ Date: 10/3/16

PASS CRITERIA: removable alpha and beta / gamma contamination less than Minimum Detectable Activity (MDA)

Radiological Leak Test: ☑ PASS ☐ FAIL

PM or Task Lead Signature: ________ Date: 10/3/16

WARNING:

The irradiation target material has been loaded into an inner housing designed to contain radioactive material, and placed into a stainless steel outer housing and welded.

THE IRRADIATION TARGET IS NOT A SEALED SOURCE. Precautions need to be taken to prevent damaging the target housing, and the housing should periodically be surveyed to confirm the housing has not been damaged or degraded.

Figure 13. Traveler sheet for target #3

NSD Irradiation Target Traveler Sheet

NSD Irradiation Target Identification Number: 67655-25-Pu4 RMT 90065

Visual inspection of weld (Step 7.4.2):

Weld Supervisor Signature: Date: 9/29/2016

1[st] Radiological Leak Test (Step 7.5.1):

Removable alpha contamination: <12 dpm / 100 cm^2

Removable beta/gamma contamination: <100 dpm / 100 cm^2

RPT Signature: Date: 9/29/2016

2[nd] Radiological Leak Test (Step 7.5.2):

Removable alpha contamination: <13 dpm / 100 cm^2

Removable beta/gamma contamination: <100 dpm / 100 cm^2

RPT Signature: Date: 10/3/16

PASS CRITERIA: removable alpha and beta / gamma contamination less than Minimum Detectable Activity (MDA)

Radiological Leak Test: ☑ PASS ☐ FAIL

PM or Task Lead Signature: Date: 10/3/16

WARNING:

The irradiation target material has been loaded into an inner housing designed to contain radioactive material, and placed into a stainless steel outer housing and welded.

THE IRRADIATION TARGET IS NOT A SEALED SOURCE. Precautions need to be taken to prevent damaging the target housing, and the housing should periodically be surveyed to confirm the housing has not been damaged or degraded.

Figure 14. Traveler sheet for target #4

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