# Characterization and Certification of Plutonium Isentropic Compression Experiment (Pu-ICE) Containment Vessels for Shipment and Emplacement at the Waste Isolation Pilot Plant (WIPP). LA-UR-13-26085, SAND 2013-9625C– 14046

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## ABSTRACT

The Plutonium Isentropic Compression Experiments (Pu-ICE) provide a scientifically based approach to stock pile stewardship and involve coordinated tasks to be completed between Los Alamos National Laboratory (LANL) and Sandia National Laboratory (SNL). Once the experiments are complete the containment vessels used during the experiments are shipped back to LANL for characterization, certification and shipment to the WIPP. This process takes the unique waste stream matrix (1200 kg of steel per drum) into consideration. An innovative approach to address these conditions has been proposed that involves direct analysis of plutonium (Pu) targets prior to shipment to SNL and visual examination (VE) from the point the targets are fabricated to the time the containment vessels are packaged for direct shipment to the Waste Isolation Pilot Plant (WIPP) from SNL.

## INTRODUCTION

With the discontinuation of above- and below-ground testing of nuclear weapons, LANL and SNL entered into a Memorandum of Understanding (MOU) defining roles and responsibilities for a cooperative project in support of a science-based approach to stockpile stewardship.[1] The U.S. Department of Energy (DOE) and SNL developed a Z pulsed-power generator (Z-Machine) to conduct laboratory experiments to verify the reliability of the nation's nuclear stockpile and LANL defined plans for experiments for the Z-machine. LANL provides the plutonium (Pu) targets, SNL conducts the experiments on the targets, and the resulting post-experiment containment vessels become transuranic (TRU) waste. The TRU waste is eligible for disposal at the WIPP.

[1] McMillan, Charles F. and Hommert, Paul, Memorandum of Understanding between Sandia Corporation and Los Alamos National Security, LLC, August 2011, Sandia MOU 11-S-560 and LANL MOU-0066, Los Alamos National Laboratory, Los Alamos , New Mexico, and Sandia National Laboratories, Albuquerque, New Mexico.

As TRU waste, the post-experiment containment vessels, Vent Tank, Upper Containment Chamber (UCC), Ultra-Fast Closure Valve (UCV) and the Load Assembly (LA), must be sufficiently characterized and certified before they can be shipped to the WIPP. This requires collection of acceptable knowledge (AK) and certification of this waste in accordance with the WIPP Waste Acceptance Criteria (WAC)[2] for radionuclides and in accordance with the WIPP Hazardous Waste Permit to identify the waste matrix, absence of prohibited items and any hazardous constituents in the waste stream.[3]

## PU-ICE PROCESS

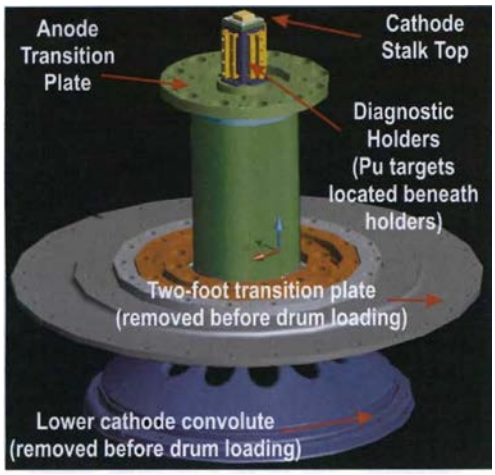


**Fig. 1: Targets Installed in the Transition Plate as Loaded into the UCC.**

LANL fabricates Pu targets and then ships them to SNL under Material Control and Accountability (MC&A) protocols established between the two laboratories [4] . The targets are received by SNL and are then loaded into the Z Machine under MC&A

[2] DOE Carlsbad Field Office (CBFO*), Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant*, Revision 7.4, April 22, 2013, DOE/WIPP-02-3122, Department of Energy Carlsbad Field Office, Carlsbad, New Mexico.

[3] CBFO, Waste Isolation Pilot Plant Hazardous Waste Permit, Attachment C, Waste Analysis Plan, March 13, 2013, Department of Energy Carlsbad Field Office, Carlsbad, New Mexico.

[4] Los Alamos National Laboratory, *Preparing Samples for Materials Characterization,* MST16-WI-649, R0-EXT6, January 14, 2010, and *Panel Preparation for Z Experiments,* MST16-EWD-WI-606, Revision 1, December 12, 2005, Los Alamos National Laboratory, Los Alamos, New Mexico. And Sandia National Laboratories, MBA 160 Custodian Procedure.

control. Throughout this process, LANL owns the Pu used in the Pu-ICE experiment and maintains ownership of the associated waste. Once the containment vessel has been used, it is temporarily stored at SNL in WIPP-approved containers under MC&A control until safeguards are terminated and the waste is shipped to LANL.

After fabrication of the Pu targets, LANL reports the isotopic composition of each target to SNL. This is tracked as part of MC&A both at LANL and SNL. Once the isotopic composition is reported to SNL, the targets are shipped to SNL, where the parts are assembled in the Z-machine. Figure 1 shows the LA prior to assembly with the other components of the containment vessel.

The radionuclides that are loaded into the containment vessel are effectively sealed in the vessel upon loading and after the experiment is complete. The radionuclides remain unchanged in composition and content during these times. The Pu-ICE experiment consists of a high current pulse flowing through the assembly under vacuum in a few microseconds. During this experiment pressures range from 10 kilobar up to 15,000 kilobar and the operating temperatures reach from 270 to 2000 degrees Kelvin.

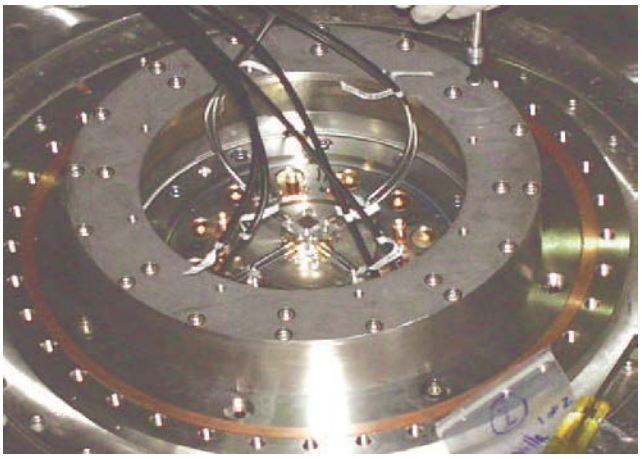

**Fig. 2: Inner Baffle during Installation of the LA.**

These conditions cause the target materials in the UCC to vaporize and the Pu is alloyed onto the UCC.[5] Figure 2 shows the inner baffle region of a containment vessel as the LA is being installed. Figure 3 shows this region after the experiment has been conducted. The Pu targets and the remaining material that has not been vaporized remains sealed in the UCC, as evidenced by the condition of the baffle region after the experiment and as verified by swipe data obtained upon completion of the experiment.[6]

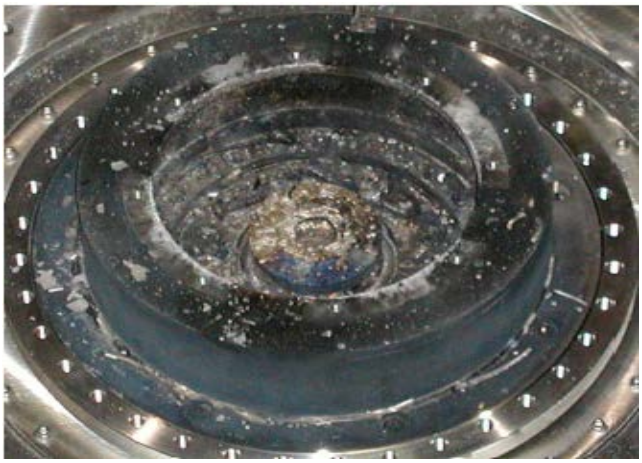

**Fig. 3: Baffle Region after Experiment is Complete.**

[5] Figures were obtained from a memo from Marcus Knudson to Alison Winstead, Re*: Condition of Special Nuclear material (SNM) post shot*, memo dated May 4, 2009, Sandia National Laboratories Albuquerque, New Mexico.

[6] Swipe data were obtained in two emails from Betty Humphrey to Beverly Crawford for all shots from 2010 to present (Re: Validation Plan both dated June 19, 2013). None of the swipe data showed any sign of radiological contamination on the outside of the containment vessel.

### Handling of Post Shot Containers

In order to address disposition of the containment vessels after completion of the experiment, LANL and SNL coordinated with the DOE-Carlsbad Field Office (CBFO) and the Central Characterization Project (CCP) for waste transportation from SNL back to LANL where the waste is characterized and ultimately shipped to the WIPP upon completion of the certification process. This work was covered by the MOU mentioned previously and a shipment plan [2] between the two laboratories. The MOU between LANL and SNL calls for MC&A, safeguards of material, termination of safeguards, shipment of the waste from SNL to LANL, and approvals by CBFO covering notifications for shipments to WIPP and ultimately, disposal at the WIPP [3]. Once the waste container is sent to LANL, waste certification proceeds under CCP's TRU waste program and ultimately the waste is shipped for disposal at the WIPP.

In 2013, discussions between LANL, SNL, and CBFO were initiated to define an approved path to allow the waste to be certified under the LANL TRU waste certification program, but be shipped directly from SNL to WIPP. This resulted in a validation plan that was presented to EPA that covers a new approach for certifying the containment vessel waste stream [4].

## WASTE STREAM COMPOSITION

The Pu-ICE containment vessels are manufactured stainless steel vessels, weighing approximately 386-454 kg each, and are composed of three sections: the load assembly (LA) containing the Pu targets; the upper containment chamber (UCC) encasing the LA, the Ultra-Fast Closure Valve (UCV), and the vent tank that sits on top of the UCC. The targets are constructed of material type 52 Pu. The Pu in these targets is well characterized in accordance with the requirements of the LANL MC&A program. The laboratory analyses performed on the targets include mass spectrometry and gamma spectroscopy.

The containment vessel waste stream consists of non-mixed metal debris that is 99% metallic. The containment vessels are fabricated to specifications by SNL qualified suppliers [5]. Table I shows the expected waste material parameters calculated for a

Table I. Example Waste Material Parameters.

| Waste Material Parameter | Weight (kg) | Percent of Total Weight |
|---|---|---|
| Iron-based Metals/Alloys | 379.8 | 97.2% |
| Aluminum-based Metals/Alloys | 0.0 | 0.0% |
| Other Metals | 9.38 | 2.4% |
| Other Inorganic materials | 0.003 | 0.00% |
| Cellulosics | 0.0 | 0.0% |
| Rubber | 0.43 | 0.10% |
| Plastics (waste materials) | 0.33 | 0.08% |
| Organic Matrix | 0.53 | 0.14% |
| Inorganic Matrix | 0.09 | 0.02% |
| Solids/Gravels | 0.0 | 0.0% |
| Steel (Packaging Materials) | 28.1 | N/A |
| Plastics (Packaging Materials) | 0.0 | N/A |

waste weight of 391kg. Conditions may vary depending on the amount of material used to meet the specifications for the vessels and removal of parts from the containment vessels during packaging. The predominant material is iron-based metal with small amounts of other metals, inorganic, organic rubber and plastic.

Table II shows the average values for Pu isotopics characterized via mass spectrometry and gamma spectroscopy for 14 test shots. The data were taken from laboratory notes

for three shots completed in 2006, [7] and from shots beginning in 2010 when memos were sent to SNL accounting for the amount of material and associated isotopics for the material being tested.[8]

The radiological composition of the targets is primarily Pu-239 ranging from 84.6 to 94. 1% by mass, Pu-240 ranging from 5.24 to 5.40 % by mass and Pu-238 from 0 to 4.69 % by mass. This isotopic content varies depending on the material used in the experiments but always within specifications.

Table II. Target Isotopes from Last 14 shots.

| Radioisotope | Average Mass (g) | Average Wt. % | Wt. % Range |
|---|---|---|---|
| Pu-238 | $4.88 \times 10^{-4}$ | 0.0129 | 0 to 4.69 |
| Pu-239 | 3.31 | 92.5 | 84.6 to 94.1 |
| Pu-240 | $2.18 \times 10^{-1}$ | 5.74 | 5.24 to 5.80 |
| Pu-241 | $5.94 \times 10^{-2}$ | 1.56 | 0.043 to 5.04 |
| Pu-242 | $1.86 \times 10^{-3}$ | 0.0490 | 0 to 0.08 |
| Am-241 | $4.91 \times 10^{-3}$ | 0.129 | 0.0423 to 0.300 |

## CHARACTERIZATION

### Non-Destructive Assay

Recent analyses performed to confirm the Pu content of 3 containment vessels overpacked in a Standard Waste Box (SWB) were run on the High Energy Neutron Counter (HENC) by CCP.[9] The HENC failed in counting the neutrons from the SWB due to high background that may be the result of spallation. Spallation occurs when large numbers of neutrons are emitted from materials (such as metals containing iron) when cosmic radiation interacts with the material. With the failure of the neutron counter, characterization was completed using the gamma spectroscopy capability of the HENC and assigned isotopics based on the debris waste stream generated at Technical Area (TA)-55 at LANL. The use of gamma spectroscopy in this way provided

[7] Humphrey, Betty email to Crawford, Beverly Re: Isotopic Information dated July 19, 2013, Weston Solutions, Albuquerque.

[8] Freibert Franz to Crawford, Beverly, Re: Shot #1, dated May 6, 2013; Re: Z Shot Data, dated May 6, 2013, Los Alamos National Laboratory, Los Alamos, New Mexico.

[9] Data for SWB 66104 is included in CCP Batch Data Report 3LANDA0134 dated 2/8/2013.

results that were 10 times too low based on the known amount of Pu in the target at the time of fabrication.

The characterization of the containment vessels presents unique problems for neutron counting. First of all, the HENC is not set up for the high metal content of the containment vessels waste stream. Secondly, if the HENC were set up the protocol would most likely take longer for count times with the amount of metal present and use software corrections for spallation. This is not the protocol currently being used to analyze debris from TA-55 at LANL. Finally, in order to set up the system for this unique matrix, a representative Performance Demonstration Program sample must be constructed and run on the HENC after it is calibrated with a matrix similar to that of the UCC.

### VE on Containment Vessels

The basic compositional information for the experiment is documented in Generator Knowledge [5] and is included in AK for the waste stream. These data are then certified by confirmation using real-time radiography or visual examination (VE). These methods provide a record that the composition of the waste is correctly identified in the AK and that no prohibited items are included in the final certified package.

Recently, a VE expert was brought in to view the process. It was determined that VE can be used throughout the Pu-ICE process. Therefore, VE will be performed from the point targets are fabricated until the final targets are prepared for shipment to the WIPP in order to confirm the composition of the waste and the absence of prohibited items. In this way, VE can be obtained without the need to return the packaged containment vessels to LANL.

## NEW APPROACH TO CERITIFICATION

An innovative, WIPP compliant, approach is needed to eliminate the need for multiple shipments to ensure characterization is complete. This approach should also provide better conditions for radiological certification of the waste without impacting the time limits required to support the Pu-ICE and reduce cost in certifying and shipping the waste to the WIPP.

The WIPP WAC allows for the use of AK under specific situations for certification of waste. In the language of the WIPP WAC:

> “Existing radioassay data collected prior to the implementation of a quality assurance program pursuant to 40 CFR § 194.22(a)(1) may only be qualified in

accordance with an alternate methodology that is approved by CBFO and employs one or more of the following methods:

- Peer review in accordance with NUREG-1297,
- Corroborating data,
- Confirmatory testing (i.e. testing on a representative sub-population of payload containers within a waste stream), or
- Demonstrating the equivalence of an alternate QA program [Section A.1 Para. 4]

SNL does not have a quality assurance program pursuant to 40 CFR § 194.22(a)(1). However, the established Central Characterization Program (CCP) at LANL can be used to confirm the Pu in the target prior to shipment to SNL. This data could then be used as confirmatory testing for a sub-population of 10 containers. Ultimately, the Pu content reported on the MC&A records for the target and the presence of other radionuclides, specifically Am-241, could be shown to be adequate for reporting the radionuclide content of each of the containers in the containment vessel waste stream.

There are several considerations that support early characterization of the targets in the LA before they are placed in the containment vessels. These considerations are:

- The waste is newly-generated and can be characterized and documented as it is generated.
- The specifications and material inputs for the containment vessels are well documented and do not change significantly from container to container.
- The Pu targets are fabricated from special nuclear material, the isotopic composition of which is well known and changes very little from one sample to the next.
- All work is documented under a strict security and quality assurance program.
- LANL has prepared a Generator Knowledge Report [5] with attachments for each experiment that includes procedures, calibrations, video recordings, weight of materials, swipe data and other AK required information.

The approach to certifying the radionuclide content of the containment vessel waste stream involves two phases. The first phase is to confirm the data from the laboratory analysis for the Pu used in the targets. This will be accomplished by assaying the targets at LANL before shipment to the WIPP. Confirmatory testing will continue for

approximately 2 years or until data from 10 shots has been obtained. The data will be used to confirm the laboratory data. The results of the data confirmation will be presented to the EPA to determine if the laboratory MC&A data can be used to certify the radiological contributions for the waste stream.

By following this path, the radionuclides in the targets can be assayed without interference from large amounts of steel from the containment vessels and radiological data are available for certification of the waste after the containment vessels are packaged at SNL. Once confirmatory data has been collected and evaluated, the handling of Pu targets can be further expedited by the use of MC&A data. Studies of target degradation have shown that targets are unusable for the Pu-ICE in as little as two weeks [6] and therefore, any time lost in confirmatory testing should be minimized in order not to impact the experiment.

## CONCLUSION

A new approach to characterize and certify radioisotopes and to ensure prohibited items is not present in the PU-ICE containment vessel waste stream has been presented here. Using this approach, the interference problems from spallation resulting from large amounts of metal in the containment vessels are eliminated during neutron counting. The addition of VE throughout the process makes certification data available without the need to return the containment vessels to LANL. Therefore, containment vessels can be shipped directly from SNL using certification data and certified shippers from CCP to send the waste to the WIPP. In so doing, the process for shipping containment vessels for Pu-ICE are efficiently and cost effectively shipped to the WIPP directly from SNL.

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3. Goyal, Kapil K., Humphrey, Betty J., Gluth, Jeffry, French, David M., *Disposition of Transuranic Residues from Plutonium Isentropic Compression Experiment (Pu-ICE) Conducted at Z-Machine,* LAUR-10-04699, Proceedings of the 13th International Conference on Environmental Remediation and Radioactive Waste Management, ICEM 2010 October 3-7, 2010, Tsukuba, Japan.

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