LA-UR-12-23255

Approved for public release; distribution is unlimited.

Title: CMR Shuffler System: Passive Mode Calibration and Certification Report

Author(s): Frame, Katherine C.
Gomez, Cipriano D.
Salazar, William R.
Mayo, Douglas R.
Vigil, Georgiana M.
Crooks, William J.
Stange, Sy

Intended for: Report

Disclaimer:

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

CMR Shuffler System: Passive Mode Calibration and Certification Report

Katherine C. Frame

William J. Crooks

Cipriano D. Gomez

Douglas R. Mayo

William R. Salazar

Sy Stange

Georgiana M. Vigil

Los Alamos National Laboratory

LA-UR-12-XXXXX

CMR Shuffler System in Passive Mode

Executive Summary:	3
Introduction	4
MC&A Measurements	5
Shuffler System	6
Shuffler Detector Parameters Summary	7
Calibrations Sources and Standards	8
Measurement Control Source	8
Calibration Standards	9
Passive Mode CMR Shuffler system Calibration	10
Position bias	12
Summary and Conclusions	12
Acknowledgements	13
References	13
Appendix A	14

EXECUTIVE SUMMARY:

Los Alamos National Laboratory has a number of spherical confinement vessels (CVs) remaining from tests involving nuclear materials. These vessels have an inner diameter of 6 feet with 1 to 2 inch thick steel walls. The goal of the Confinement Vessel Disposition (CVD) project is to remove debris and reduce contamination inside the vessels. As debris is removed from the vessels, material will be placed in waste drums. Far-field gamma ray assay will be used to determine when a drum is nearing a ^{239}Pu equivalent mass of less than 200 g. The drum will then be assayed using a waste drum shuffler operated in passive mode using a neutron coincidence counting method for accountability.

This report focuses on the testing and calibration of the CMR waste drum shuffler in passive mode operation. Initial testing was performed to confirm previously accepted measurement parameters. The system was then calibrated using a set of weapons grade Pu (WG Pu, $^{239}\text{Pu} > 93\%$) oxide standards placed inside a 55 gallon drum. The calibration data ranges from Pu mass of 0.5 g to 188.9 g.

The CMR waste drum shuffler has been tested and calibrated in passive mode in preparation for safeguards accountability measurements of waste drums containing material removed from CVs for the CVD project.

INTRODUCTION

The Confinement Vessel Disposition (CVD) project is part of a larger effort to consolidate and dispose of non-programmatic special nuclear materials (SNM) at Los Alamos National Laboratory (LANL). The charter of the CVD project is to remove debris and contamination from vessels that were used in 1970s-era explosives experiments. These spherical vessels have a 6-ft (1.83-m) inner diameter and 1- or 2-in (25.4- or 50.8-mm) thick steel walls, weighing upwards of 7500 lbs (3300-kg). Figure 1 provides a photographic example of several of these vessels in storage at LANL. Each vessel contains ~500 lb of radioactive debris and may include up to 11 lb (5-kg) ^{239}Pu equivalent in the form of actinide metals and oxides mixed in a matrix of metal (steel, brass, aluminum), beryllium, lead, powdered silica, graphite, electrical wires and other hardware. Figure 2 gives an example of the debris inside of a vessel prior to and after cleanout from a previous campaign in which hand tools were used to remove debris and reduce contamination in the vessel.

Figure 1. Several confinement vessels pictured in their current storage location at LANL.

Figure 2. Inside view showing confinement vessel before (left) and after (right) cleanout.

The current CVD project has included equipment and procedural upgrades for the removal of debris to include a robotic arm and glove box workstation in order to improve contamination control and reduce worker risk during cleanout. Figure 3 shows an artist's conception of the confinement vessel during the cleanout process. While the vessel is supported on a stand, the glove box workstation will be connected through one port on the vessel and the robotic arm will be inserted through another port. The robotic arm can be fitted with different attachments for cutting up large metal pieces, clearing debris, and scrubbing the walls of the vessel. Lights and cameras may also be attached for inspection during and after the cleanout. Within the glove box,

CMR Shuffler System in Passive Mode

hand tools may be used to further reduce the size of metal pieces. Material that can be used in programmatic applications will be recovered and non-programmatic materials will be bagged out and placed in waste drums for disposition.

Figure 3. Configuration of the confinement vessel, glove box workstation, and robotic arm during the performance of cleanout operations.

MC&A Measurements

We were tasked to design, develop, implement, and certify nondestructive assay systems [1] that would meet all of the various stages associated with the disposition of materials from the confinement vessels (CVs). In order to meet the safeguards requirements, a set of measurement methods have been accepted for use on this project. A combination of neutron and gamma ray measurements will be used for Materials Control and Accountability (MC&A) throughout the CVD project. Measurements will be performed in the following sequence for the disposition of each confinement vessel.

1. Gamma ray assay of the glove box workstation to determine holdup of SNM from previous confinement vessel cleanout (presumed to be zero prior to cleanout of the first CV, then some small value as the campaign progresses).
2. Neutron assay [2][3] of the confinement vessel before processing to verify and fully document the initial material inventory.
3. Far-field gamma-ray assay of port covers after removal to determine their levels of contamination.
4. Long-dwell-time gamma-ray assay of the first waste drum to establish isotopic composition of the SNM in the vessel.

CMR Shuffler System in Passive Mode

5. Far-field gamma ray assay of each waste drum to assess when the drum is reaching an amount below the maximum quantity of ^{239}Pu equivalent (< 200 g) as well as to determine the ratio of uranium to plutonium content.
6. Neutron assay of waste drums using a waste drum shuffler operated in passive mode to quantify SNM content in each drum [4][5].
7. Neutron measurement of the confinement vessel upon completion of cleanup operations to determine the residual content of SNM [3].
8. Gamma ray measurement of the glove box workstation upon completion of debris removal from the vessel to determine an ending holdup value for the workstation.

This paper focuses on the passive shuffler assay system that will be used to perform neutron coincidence counting measurements of the waste drums containing less than 200 g ^{239}Pu equivalent.

SHUFFLER SYSTEM

The shuffler is located in the nondestructive assay (NDA) laboratory (Room S103) of the Chemistry and Metallurgy Research (CMR) facility and is operated by the CMR Operations (CMR-OPS) group, NDA laboratory personnel and Nuclear Nonproliferation Division (N-Division) technical staff. Canberra Industries fabricated the shuffler, which was designed by the Safeguards Science and Technology Group (NEN-1) in the NEN-Division [6][7]. The shuffler can be used to measure items of any size and geometry up to 55-gallon drums. The shuffler is a passive/active neutron (PAN) instrument that assays plutonium (Pu) and/or uranium. The CMR Shuffler will only be used in the passive mode for neutron assay of waste drums for the CVD project.

In the passive mode, the PAN shuffler acts as a neutron coincidence counter (NCC) for measuring spontaneously fissioning isotopes, ^{238}Pu , ^{240}Pu , and ^{242}Pu . Pu is assayed in this mode by counting multiples of coincident neutrons from the spontaneously fissioning even Pu isotopes using the International Neutron Coincidence Counting (INCC) software. In the active mode (which will not be enabled), the shuffler interrogates (induces fissions in) the fissile material (for example, uranium) by neutrons to induce fissions and counts the resulting delayed neutrons from the precursors (fission fragments). The interrogating neutrons come from a ^{252}Cf (californium) source which is "shuffled" a number of times from a shielded position to the assay chamber and back by a computer-controlled stepping motor. Banks of polyethylene-moderated helium (^3He) detectors surround the assay chamber and are used to count neutrons in either mode of operation.

The Shuffler hardware includes a shuffler body and an electronics rack. A photograph of the CMR Shuffler body is shown in Figure 2. The detector consists of eight banks of ^3He tubes embedded in high-density polyethylene moderator. Six detector banks surround an assay chamber with a rotating floor. More detail can be found in [6] and [7].

Figure 4. Photograph of the CMR waste drum shuffler body.

Shuffler Detector Parameters Summary

The passive shuffler detector parameters are shown in Table 1. The die-away, predelay, gate width and dead-time parameters were taken from the parameter settings previously stored in the system. Measurements were performed to confirm the die-away and gate width settings, and the efficiency was also measured. During initial testing, unusual fluctuations in background count rates were observed in the bottom bank of detectors. The decision was made to disable this bank to reduce measurement uncertainty due to these fluctuations. This bank is also disabled in the TA-55 shuffler.

Table 1. Shuffler detector parameters.

Die-away	$\tau[\mu\text{s}]$	71
Predelay	$P[\mu\text{s}]$	4.5
Gate Width	$GW[\mu\text{s}]$	90
Efficiency	ϵ	14.7%
Dead-time	$A[\mu\text{s}]$	0.4383
	$B[\mu\text{s}^2]$	0.1461
High Voltage	HV (V)	1680

CMR Shuffler System in Passive Mode

The die-away time was measured using a ^{252}Cf source and the following method [9]. The probability (dp) for capturing a neutron in a ^3He tube in a time dt is given approximately by

$$dp = \frac{1}{\tau} \exp(-t / \tau) \cdot dt ,$$

where t is the time from emission to capture and τ is the detector die-away time. The probability (p) of detecting a neutron in the coincidence gate is thus

$$p = \exp(-P / \tau) \cdot [1 - \exp(-G / \tau)] ,$$

where P is the predelay and G is the gate length. Using the above equation, the ratio of the doubles coincident counts at two different gate widths, 128 μs and 64 μs , is given by

$$\frac{R_{128}}{R_{64}} = \frac{1 - \exp(-128 / \tau)}{1 - \exp(-64 / \tau)} .$$

Solving this equation for τ gives

$$\tau = \frac{-64}{\ln\left(\frac{R_{128}}{R_{64}} - 1\right)} .$$

The die-away time measured using this method was found to be 68.3 μs . An optimal gate-width has been shown to be $1.27 \cdot \tau = 87 \mu\text{s}$ [1]. These values are comparable to the parameter settings of 71 μs die-away and 90 μs gate width.

The efficiency was measured to be 14.7% by placing a ^{252}Cf source on a stand in the center of the assay chamber. The efficiency was calculated as the ratio of the measured singles rate to the known yield of the source. NIST Source traceability measurements were performed for the ^{252}Cf source (ID A7-258) using the SNAP-III detector on December 22nd and 23rd of 2009. The source characteristics were originally reported to be 500 μCi on August 15th of 2002. Measurements on December 21st of 2009 revealed a neutron yield of 2.81×10^5 n/s. The value previously stored was 15.4%. With the bottom bank enabled, we measured an efficiency of 15.2%.

CALIBRATIONS SOURCES AND STANDARDS

Measurement Control Source

The ^{252}Cf source, A7-258, that was used to measure the efficiency was also used for measurement control during calibration of the system.

CMR Shuffler System in Passive Mode

Calibration Standards

A set of weapons grade Pu (WGPu, $^{239}\text{Pu} > 93\%$) standards were used to calibrate the system. The PDP and RANT series contain high purity plutonium oxide (PuO_2) dispersed in diatomaceous earth. Certificates of content and traceability are included in Appendix A with the exception of the PDP30.0Pu2.0mm standard. This standard contains large particle (2.0mm) oxide mixed with the diatomaceous earth and we have included a Los Alamos interoffice memorandum, which gives the Pu mass and isotopic composition of this standard. Table 2 contains source identifications along with the declared Pu mass and effective ^{240}Pu ($^{240}\text{Pu}_{\text{eff}}$) mass on the date that calibration data were collected. The calibration data ranges from Pu masses of 0.5 g to 188.9 g ($^{240}\text{Pu}_{\text{eff}}$ masses of 0.03 g to 13 g).

Table 2 Calibration Standards.

Source ID	Measurement Date	$^{240}\text{Pu}_{\text{eff}}$ mass (g)	Pu Mass (g)
PDP0.50Pu	10/18/11	0.03	0.48
PDP1.0Pu	10/19/11	0.07	0.95
PDP3.0Pu	10/19/11	0.18	3.04
PDP5.0Pu	10/17/11	0.34	4.99
PDP10.0Pu	10/14/11	0.61	10.04
PDP5.0Pu and PDP10.0Pu	10/18/11	0.95	15.04
PDP20.0Pu	10/17/11	1.37	20.00
PDP30.0Pu2.0mm	10/17/11	2.08	30.26
PDP30.0Pu2.0mm and PDP10.0Pu	10/18/11	2.69	40.31
RANT50-1	11/28/11	3.43	49.96
PDP30.0Pu2.0mm and PDP20.0Pu	10/18/11	3.45	50.26
PDP30.0Pu2.0mm, PDP20.0Pu and PDP10.0Pu	10/18/11	4.06	60.31
RANT50-1 and RANT50-2	11/28/11	6.86	99.89
RANT50-1, RANT50-2 and RANT50-3	11/28/11	10.29	149.85
RANT50-1, RANT50-2, RANT50-3, PDP20.0Pu and PDP5.0Pu	11/28/11	12.00	174.81
RANT50-1, RANT50-2, RANT50-3, PDP1.0Pu, PDP3.0Pu, PDP5.0Pu, PDP10.0Pu and PDP20.0Pu	11/29/11	12.86	188.88

PASSIVE MODE CMR SHUFFLER SYSTEM CALIBRATION

Verification measurements will be performed on 55-gallon waste drums containing material removed from CVs. High resolution gamma ray assay will be used to measure the relative isotopic content of the material and far-field gamma-ray assay will be used to determine when a drum is nearing a ^{239}Pu equivalent mass of less than 200 g.

The calibration is based on doubles coincidence counting. Waste drums from vessels containing beryllium will require expert review. Beryllium has a very high cross-section for (α, n) interactions, and these neutrons may induce fissions that will increase the doubles rate. Additional review will be needed to estimate a correction to the doubles rate that can then be used to determine the mass.

Prior to performing calibration measurements, background measurements were performed with an empty assay chamber. The standards were then placed inside of a 55-gallon drum with positioning tubes that were fabricated to hold these types of standards at various positions within the drum. There are 3 tubes, aligned vertically within the drum. Tube 1 is at the center, Tube 2 is approximately half way between the center and the outer edge of the drum and Tube 3 is near the outer edge of the drum. Each tube has 3 vertical positions within it. For cases where a single standard was measured, the standards were positioned in the vertical center of Tube 2. For cases where 2 and 3 standards were measured together, the standards were positioned in the vertical centers of Tube 1 and Tube 2 (for 2 standards) or Tube 1, Tube 2, and Tube 3 (for 3 standards). In cases where 5 standards were measured, the center and upper positions were used, and in the case where 8 standards were measured all positions except for the bottom of Tube 3 was used.

Figure 5 shows the doubles coincident count rate as a function of $^{240}\text{Pu}_{\text{eff}}$ mass from 0.03 g to 13 g (Pu masses of 0.03 g to 13 g). Error bars are within the polymarker. A linear fit was performed on these data and the result is also shown in Figure 5 as a straight line. The calibration curve has the functional form, $\text{Doubles Rate} = 16.12659 \times ^{240}\text{Pu}_{\text{eff}} \text{ mass}$.

Figure 5 Calibration curve, Doubles rate as a function of ²⁴⁰Pu_{eff} mass.

To verify the calibration, a combination of standards was measured that was not used for calibration. The RANT50-1, RANT50-2, and PDP20.0Pu standards were placed in the vertical centers of Tubes 1, 2, and 3 of the drum. A verification measurement was performed and the results are shown in Table 3. The difference between the declared and assay masses agree well within the uncertainty.

Table 3. Verification results for a combined Pu mass of 120 g.

Assay ²⁴⁰ Pu _{eff} Mass (g) :	8.200 ± 0.068
²⁴⁰ Pu _{eff} (%):	6.866
Assay Pu Mass (g):	119.437 ± 0.996
Declared ²⁴⁰ Pu _{eff} Mass (g):	8.232
Declared Pu Mass (g):	119.895
Declared – Assay Pu Mass (g):	0.458 ± 0.996
Declared – Assay Pu Mass (%):	0.382 ± 0.830

POSITION BIAS

When a waste drum is assayed, the exact distribution of material within the drum will be unknown. As described above, the calibration was performed with the material distributed near the vertical center of the drum. In the worst-case scenario, all of the material may be located at one particular location and the assay result may be biased. To estimate an uncertainty, measurements were performed with one of the 50 g standards (RANT50-1) placed in multiple locations in the drum and the doubles rate was compared to the doubles rate when the source was placed in the vertical center of Tube 2. The largest deviation was found with the source positioned in the bottom of Tube 1. With the bottom detector bank disabled, this location has the poorest solid angle coverage. Unfortunately, it is also likely that material will settle at the bottom of the drum.

The doubles rate for the source at the nominal location is 1.36 times that of the doubles rate for the source located at the bottom of the drum. If we were to assay a waste drum assuming a distribution similar to that of the calibration measurements, but the material is actually concentrated at the bottom of the drum, we would underestimate the amount of material and there could be as much as 36% more material than the assay amount. If we were to assay a drum where the material is near the top of the drum, we would overestimate the amount by only 3.5%.

To accommodate a position bias, we will apply an asymmetric uncertainty of 36% in the positive direction and 3.5% in the negative direction. This will be added in quadrature to the assay uncertainty reported by INCC. This uncertainty will be the dominant uncertainty.

SUMMARY AND CONCLUSIONS

The CMR waste drum shuffler will be operated in passive mode to perform assay measurements of waste drums containing material that has been removed from CVs. The drums will be measured using far field gamma-ray assay to ensure they are below the 200 g ^{239}Pu equivalent mass limit prior to shuffler assay.

The system has been calibrated for $^{240}\text{Pu}_{\text{eff}}$ masses from 0.03 g to 13 g (corresponding to 0.5 g to 189 g WGPu). The standards used for calibration contain high purity PuO_2 mixed with diatomaceous earth. The calibration measurements were performed with standards located near the vertical center of a 55 gallon drum configured for positioning these standards in a variety of locations. The calibration was verified by performing assay measurements on a combination of standards not used for calibration. Assayed masses agreed with declared masses within 1 σ , statistical error.

The systematic uncertainty due to position bias was estimated by placing a standard at multiple locations within the drum used for calibration. It was found to have the largest deviation at the bottom of the drum where is also most likely that material will settle prior to assay. To accommodate a position bias, an asymmetric uncertainty of +36% and -3.5% will be added in quadrature to the assay uncertainty reported by INCC.

One limitation to the validity of the calibration curve is the assay of material containing beryllium, and this will require expert review.

CMR Shuffler System in Passive Mode

The CMR waste drum shuffler has been tested and calibrated in preparation for and safeguards measurements of waste drums containing material removed from CVs during cleanout.

Acknowledgements

We gratefully acknowledge many helpful hours spent in collaboration with our colleagues who support the Chemistry and Metallurgy Research Facility (CMR) operations. In addition, we acknowledge financial support from DOE.

REFERENCES

- [1] D. Reilly et al., *Passive Nondestructive Assay of Nuclear Materials*, NUREG/CR-5550, LA-UR-90-732, United States Nuclear Regulatory Commission, March 1991.
- [2] M. Krick, “²⁴⁰Pu-effective Mass Formula for Coincidence Counting of Plutonium with Shift Register Electronics,” in “Nuclear Safeguards Program Status Report, May-August 1977,” J. L. Sapir, Comp., Los Alamos Scientific Laboratory report LA-7030-PR (1977), p. 16.
- [3] K. Frame et al., “Confinement Vessel Assay System Design and Implementation Report”, Los Alamos National Laboratory internal document, 2012.
- [4] P. Rinard, “Application Guide to Shufflers,” Los Alamos National Laboratory document LA-13819-MS, September 2001.
- [5] P. Rinard, “Shuffler Instruments for the Nondestructive Assay of Fissile Materials,” Los Alamos National Laboratory document LA-12105, May 1991.
- [6] P. Rinard et al., *Hardware Manual for Waste Drum Shufflers Model JCC-92*, Canberra Industries, Meriden, CT, November 1993.
- [7] P. Rinard et al., “Los Alamos Waste Drum Shufflers Users Manual,” Los Alamos National Laboratory document LA-UR-93-3148, August 1993.
- [8] W. Harker and M. Krick, “Neutron Coincidence Counting Software for Windows,” Los Alamos National Laboratory document, LA-UR-96-360, 1996.
- [9] P. Santi et al., “Nondestructive Assay Inspector Training Course”, Los Alamos Laboratory Document LA-UR-06-5614, 2006.
- [10] N. Ensslin et al., “Application Guide to Neutron Multiplicity Counting,” Los Alamos National Laboratory document LA-13422-M, 1998.

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard PDPO.10Pu

This NDA standard contains high purity plutonium dioxide dispersed in diatomaceous earth. Quantitative information and uncertainties on the nuclear content of this WRM are listed below. Complete information regarding the Pu and Am content, Pu isotopic ratios, chemical composition, elemental impurities, containment, and fabrication procedures are retained in files by the Los Alamos National Laboratory Inorganic Elemental Analysis Group.

NUCLEAR MATERIAL CONTENT

The nuclear material content and total alpha activity for this standard are listed below. Also listed are overall uncertainty estimates at 95% confidence intervals (CI) for each component.

Component	Quantity or Value	95 % CI
PuO ₂ in standard	0.11587 grams	± 0.00023 grams ^(a)
Pu Assay	87.915 % of PuO ₂	± 0.065 % ^(b)
Pu in standard	0.10187 grams	± 0.00021 grams ^(c)
²⁴¹ Am Assay	979.2 µg/g PuO ₂ *	± 0.2 µg/g ^(d)
²⁴¹ Am in standard	0.1135 mg	± 0.0003 mg ^(e)
Isotopic Weight Fraction		
²³⁸ Pu	0.000151	± 0.000005 ^(f)
²³⁹ Pu	0.929661	± 0.000015 ^(f)
²⁴⁰ Pu	0.067202	± 0.000007 ^(f)
²⁴¹ Pu	0.002327*	± 0.000010 ^(f)
²⁴² Pu	0.000659	± 0.000003 ^(f)
Total Alpha Activity in standard	8.079 mCi** 2.989E+08 Bq	± 0.023 mCi ^(g) ± 9E+05 Bq ^(g)

*Measured value on 10/2/96

** Effective 10/2/96

 Bill J. McKerley, Group Leader
 Inorganic Elemental Analysis

 Robert S. Marshall, Project Manager
 NDA WRM Production

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard PDP0.50Pu

This NDA standard contains high purity plutonium dioxide dispersed in diatomaceous earth. Quantitative information and uncertainties on the nuclear content of this WRM are listed below. Complete information regarding the Pu and Am content, Pu isotopic ratios, chemical composition, elemental impurities, containment, and fabrication procedures are retained in files by the Los Alamos National Laboratory Inorganic Elemental Analysis Group.

NUCLEAR MATERIAL CONTENT

The nuclear material content and total alpha activity for this standard are listed below. Also listed are overall uncertainty estimates at 95% confidence intervals (CI) for each component.

<u>Component</u>	<u>Quantity or Value</u>	<u>95 % CI</u>
PuO ₂ in standard	0.55019 grams	± 0.00087 grams ^(a)
Pu Assay	87.915 % of PuO ₂	± 0.065 % ^(b)
Pu in standard	0.48370 grams	± 0.00077 grams ^(c)
²⁴¹ Am Assay	979.2 µg/g PuO ₂ *	± 0.2 µg/g ^(d)
²⁴¹ Am in standard	0.5387 mg	± 0.0016 mg ^(e)
Isotopic Weight Fraction		
²³⁸ Pu	0.000151	± 0.000005 ^(f)
²³⁹ Pu	0.929661	± 0.000015 ^(f)
²⁴⁰ Pu	0.067202	± 0.000007 ^(f)
²⁴¹ Pu	0.002327*	± 0.000010 ^(f)
²⁴² Pu	0.000659	± 0.000003 ^(f)
Total Alpha Activity in standard	38.36 mCi** 1.419E+09 Bq	± 0.11 mCi ^(g) ± 4E+06 Bq ^(g)

*Measured values on 10/2/96

** Effective 10/2/96

 Bill J. McKerley, Group Leader
 Inorganic Elemental Analysis

 Robert S. Marshall, Project Manager
 NDA WRM Production

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard PDP1.0Pu

This NDA standard contains high purity plutonium dioxide dispersed in diatomaceous earth. Quantitative information and uncertainties on the nuclear content of this WRM are listed below. Complete information regarding the Pu and Am content, Pu isotopic ratios, chemical composition, elemental impurities, containment, and fabrication procedures are retained in files by the Los Alamos National Laboratory Inorganic Elemental Analysis Group.

NUCLEAR MATERIAL CONTENT

The nuclear material content and total alpha activity for this standard are listed below. Also listed are overall uncertainty estimates at 95% confidence intervals (CI) for each component.

Component	Quantity or Value	95 % CI
PuO ₂ in standard	1.08130 grams	± 0.00163 grams ^(a)
Pu Assay	87.915 % of PuO ₂	± 0.065 % ^(b)
Pu in standard	0.95062 grams	± 0.00144 grams ^(c)
²⁴¹ Am Assay	979.2 µg/g PuO ₂ *	± 0.2 µg/g ^(d)
²⁴¹ Am in standard	1.059 mg*	± 0.003 mg ^(e)
Isotopic Weight Fraction		
²³⁸ Pu	0.000151	± 0.000005 ^(f)
²³⁹ Pu	0.929661	± 0.000015 ^(f)
²⁴⁰ Pu	0.067202	± 0.000007 ^(f)
²⁴¹ Pu	0.002327*	± 0.000010 ^(f)
²⁴² Pu	0.000659	± 0.000003 ^(f)
Total Alpha Activity in standard	75.39 mCi** 2.789E+09 Bq	± 0.22 mCi ^(g) ± 8E+06 Bq ^(g)

*Measured value on 10/2/96

** Effective 10/2/96

 Bill J. McKerley, Group Leader
 Inorganic Elemental Analysis

 Robert S. Marshall, Project Manager
 NDA WRM Production

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard PDP3.0Pu

This NDA standard contains high purity plutonium dioxide dispersed in diatomaceous earth. Quantitative information and uncertainties on the nuclear content of this WRM are listed below. Complete information regarding the Pu and Am content, Pu isotopic ratios, chemical composition, elemental impurities, containment, and fabrication procedures are retained in files by the Los Alamos National Laboratory Inorganic Elemental Analysis Group.

NUCLEAR MATERIAL CONTENT

The nuclear material content and total alpha activity for this standard are listed below. Also listed are overall uncertainty estimates at 95% confidence intervals (CI) for each component.

<u>Component</u>	<u>Quantity or Value</u>	<u>95 % CI</u>
PuO ₂ in standard	3.4648 grams	± 0.0029 grams ^(a)
Pu Assay	87.827 % of PuO ₂	± 0.072 % ^(b)
Pu in standard	3.0430 grams	± 0.0026 grams ^(c)
²⁴¹ Am Assay	766.7 µg/g PuO ₂ *	± 2.2 µg/g ^(d)
²⁴¹ Am in standard	2.656 mg	± 0.0075 mg ^(e)
Isotopic Weight Fraction		
²³⁸ Pu	0.000145	± 0.000008 ^(f)
²³⁹ Pu	0.937614	± 0.000013 ^(f)
²⁴⁰ Pu	0.059445	± 0.000005 ^(f)
²⁴¹ Pu	0.002237*	± 0.000008 ^(f)
²⁴² Pu	0.000559	± 0.000004 ^(f)
Total Alpha Activity in standard		
	234.7 mCi**	± 0.8 mCi ^(a)
	8.682E+09 Bq	± 3E+07 Bq ^(a)

*Measured value on 7/15/95

** Effective 7/15/95

 Bill J. McKerley, Group Leader
 Inorganic Elemental Analysis

 Robert S. Marshall, Project Manager
 NDA PDP WRM Production

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard PDP5.0Pu

This NDA standard contains high purity plutonium dioxide dispersed in diatomaceous earth. Quantitative information and uncertainties on the nuclear content of this WRM are listed below. Complete information regarding the Pu and Am content, Pu isotopic ratios, chemical composition, elemental impurities, containment, and fabrication procedures are retained in files by the Los Alamos National Laboratory Inorganic Elemental Analysis Group.

NUCLEAR MATERIAL CONTENT

The nuclear material content and total alpha activity for this standard are listed below. Also listed are overall uncertainty estimates at 95% confidence intervals (CI) for each component.

Component	Quantity or Value	95 % CI
PuO ₂ in standard	5.6895 grams	± 0.0088 grams ^(a)
Pu Assay	87.915 % of PuO ₂	± 0.065 % ^(b)
Pu in standard	5.0020 grams	± 0.0077 grams ^(c)
²⁴¹ Am Assay	979.2 µg/g PuO ₂ *	± 0.2 µg/g ^(d)
²⁴¹ Am in standard	5.571 mg	± 0.016 mg ^(e)
Isotopic Weight Fraction		
²³⁸ Pu	0.000151	± 0.000005 ^(f)
²³⁹ Pu	0.929661	± 0.000015 ^(f)
²⁴⁰ Pu	0.067202	± 0.000007 ^(f)
²⁴¹ Pu	0.002327*	± 0.000010 ^(f)
²⁴² Pu	0.000659	± 0.000003 ^(f)
Total Alpha Activity in standard		
	396.7 mCi**	± 1.1 mCi ^(g)
	1.468E+10 Bq	± 4E+07 Bq ^(g)

*Measured value on 10/2/96

** Effective 10/2/96

B. J. McKerley, Group Leader
Inorganic Elemental Analysis

Robert S. Marshall, Project Manager
NDA WRM Production

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard PDP10.0Pu

This NDA standard contains high purity plutonium dioxide dispersed in diatomaceous earth. Quantitative information and uncertainties on the nuclear content of this WRM are listed below. Complete information regarding the Pu and Am content, Pu isotopic ratios, chemical composition, elemental impurities, containment, and fabrication procedures are retained in files by the Los Alamos National Laboratory Inorganic Elemental Analysis Group.

NUCLEAR MATERIAL CONTENT

The nuclear material content and total alpha activity for this standard are listed below. Also listed are overall uncertainty estimates at 95% confidence intervals (CI) for each component.

<u>Component</u>	<u>Quantity or Value</u>	<u>95 % CI</u>
PuO ₂ in standard	11.4570 grams	± 0.0096 grams ^(a)
Pu Assay	87.827 % of PuO ₂	± 0.072 % ^(b)
Pu in standard	10.0624 grams	± 0.0085 grams ^(c)
²⁴¹ Am Assay	766.7 µg/g PuO ₂ *	± 2.2 µg/g ^(d)
²⁴¹ Am in standard	8.784 mg *	± 0.0247 mg ^(e)
Isotopic Weight Fraction		
²³⁸ Pu	0.000145	± 0.000008 ^(f)
²³⁹ Pu	0.937614	± 0.000013 ^(f)
²⁴⁰ Pu	0.059445	± 0.000005 ^(f)
²⁴¹ Pu	0.002237 *	± 0.000008 ^(f)
²⁴² Pu	0.000559	± 0.000004 ^(f)
Total Alpha Activity in standard		
	775.9 mCi **	± 2.8 mCi ^(g)
	2.871E+10 Bq	± 1E+08 Bq ^(g)

* Measured value on 7/15/95

** Effective 7/15/95

 Bill J. McKerley, Group Leader
 Inorganic Elemental Analysis

 Robert S. Marshall, Project Manager
 NDA PDP WRM Production

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard PDP20.0Pu

This NDA standard contains high purity plutonium dioxide dispersed in diatomaceous earth. Quantitative information and uncertainties on the nuclear content of this WRM are listed below. Complete information regarding the Pu and Am content, Pu isotopic ratios, chemical composition, elemental impurities, containment, and fabrication procedures are retained in files by the Los Alamos National Laboratory Inorganic Elemental Analysis Group.

NUCLEAR MATERIAL CONTENT

The nuclear material content and total alpha activity for this standard are listed below. Also listed are overall uncertainty estimates at 95% confidence intervals (CI) for each component.

<u>Component</u>	<u>Quantity or Value</u>	<u>95 % CI</u>
PuO ₂ in standard	22.791 grams	± 0.034 grams ^(a)
Pu Assay	87.915 % of PuO ₂	± 0.065 % ^(b)
Pu in standard	20.037 grams	± 0.030 grams ^(c)
²⁴¹ Am Assay	979.2 µg/g PuO ₂ *	± 0.2 µg/g ^(d)
²⁴¹ Am in standard	22.32 mg*	± 0.07 mg ^(e)
Isotopic Weight Fraction		
²³⁸ Pu	0.000151	± 0.000005 ^(f)
²³⁹ Pu	0.929661	± 0.000015 ^(f)
²⁴⁰ Pu	0.067202	± 0.000007 ^(f)
²⁴¹ Pu	0.002327*	± 0.000010 ^(f)
²⁴² Pu	0.000659	± 0.000003 ^(f)
Total Alpha Activity in standard	1589 mCi** 5.880E+10 Bq	± 5 mCi ^(g) ± 2E+08 Bq ^(g)

*Measured value on 10/2/96

** Effective 10/2/96

 Bill J. McKerley, Group Leader
 Inorganic Elemental Analysis

 Robert S. Marshall, Project Manager
 NDA WRM Production

CMR Shuffler System in Passive Mode

Los Alamos

NATIONAL LABORATORY

Memorandum*Nuclear Material Technology*NMT-1 Analytical Chemistry
Standard Fabrication Team, MS G740
Los Alamos, New Mexico 87545*To/MS:* Diane Otero-Bell, NMT-4 LANL*From/MS:* Sandra L. Mecklenburg, NMT-1/G740*Phone/FAX:* (505) 665-7182/(505) 665-4737*Date:* May 2, 2000*Reference:* NMT-1-00-026

Below is information on the large particle plutonium sources which are being shipped to NMT-4. You are receiving seven items with different masses of Pu per item and different particle sizes. The label notation gives the approximate grams of Pu (i.e. PDP5.0Pu is ~5.0 g of Pu) and the particle size in millimeters (i.e. 2.0mm indicates the particle sizes are ~2.0 mm). The information listed below is being provided as a summary. Formal certificates containing error analysis will be sent separately when they are ready.

Summary information on the large particle Pu sources:

<u>WRM ID Number</u>	<u>Mass of Pu (g)</u>	<u>Mass of ²³⁹Pu (g)</u>	<u>Dose Rate (contact)</u>
PDP5.0Pu~2.5mm	5.05020	4.69604	1.3 mR/h
PDP5.0Pu2.0mm	5.07593	4.71996	1.5 mR/h
PDP5.0Pu1.0mm	4.98867	4.63882	2.0 mR/h
PDP5.0Pu0.5mm	5.01811	4.66620	2.4 mR/h
PDP0.5Pu2.0mm	0.50127	0.46611	0.3 mR/h
PDP15.0Pu2.0mm	15.67266	14.57355	4 mR/h
PDP30.0Pu2.0mm	30.30469	28.17945	6 mR/h

<u>Pu Isotope*</u>	<u>Weight %</u>
²³⁸ Pu	0.01490
²³⁹ Pu	92.98709
²⁴⁰ Pu	6.72067
²⁴¹ Pu	0.21142
²⁴² Pu	0.06592
²⁴¹ Am	1162.8 ppm
<u>Pu assay</u>	0.87778 g Pu/g PuO ₂ **

*Decay corrected to 9/28/98 **Analysis date 9/28/98

Los Alamos National Laboratory Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard Rant 50-1

This NDA standard contains high purity plutonium dioxide dispersed in diatomaceous earth. Quantitative information and uncertainties on the nuclear content of this WRM are listed below. Complete information regarding the Pu and Am content, Pu isotopic ratios, chemical composition, elemental impurities, containment, and fabrication procedures are retained in files by the Los Alamos National Laboratory CMR Analytical Chemistry Group.

NUCLEAR MATERIAL CONTENT

The nuclear material content and total alpha activity for this standard are listed below. Also listed are overall uncertainty estimates at 95% confidence intervals (CI) for each component.

<u>Component</u>	<u>Quantity or Value</u>	<u>95 % CI</u>
PuO ₂ in standard	56.9204 grams	± 0.05292 grams ^(a)
Pu Assay	87.897 % of PuO ₂ *	± 0.081 % ^(b)
Pu in standard	50.03132 grams	± 0.04652 grams ^(c)
²⁴¹ Am Assay	1140.3 µg/g PuO ₂ *	± 0.3 µg/g ^(d)
²⁴¹ Am in standard	64.89 mg*	± 0.43 mg ^(e)
Isotopic Weight Fraction		
²³⁸ Pu	0.000149	± 0.000005 ^(f)
²³⁹ Pu	0.929845	± 0.000015 ^(f)
²⁴⁰ Pu	0.067206	± 0.000007 ^(f)
²⁴¹ Pu	0.002141*	± 0.000009 ^(f)
²⁴² Pu	0.000659	± 0.000003 ^(f)
Total Alpha Activity in standard		
	3997 mCi**	± 11 mCi ^(g)
	1.479E+11 Bq	± 4E+08 Bq ^(g)

*Decay corrected value on 6/25/98 ** Effective 6/25/98

Peggy Hartley, Acting
Bill J. McKerley, Group Leader
CMR Analytical Chemistry

Robert S. Marshall
Robert S. Marshall, Project Manager
NDA WRM Production

64890 µg
50.03132 g = 1226.22 µg/g at 6/25/98

404 mg

(over)

Los Alamos National Laboratory Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard Rant 50-2

This NDA standard contains high purity plutonium dioxide dispersed in diatomaceous earth. Quantitative information and uncertainties on the nuclear content of this WRM are listed below. Complete information regarding the Pu and Am content, Pu isotopic ratios, chemical composition, elemental impurities, containment, and fabrication procedures are retained in files by the Los Alamos National Laboratory CMR Analytical Chemistry Group.

NUCLEAR MATERIAL CONTENT

The nuclear material content and total alpha activity for this standard are listed below. Also listed are overall uncertainty estimates at 95% confidence intervals (CI) for each component.

<u>Component</u>	<u>Quantity or Value</u>	<u>95 % CI</u>
PuO ₂ in standard	56.8965 grams	± 0.05309 grams ^(a)
Pu Assay	87.897 % of PuO ₂ *	± 0.081 % ^(b)
Pu in standard	50.01032 grams	± 0.04667 grams ^(c)
²⁴¹ Am Assay	1140.3 µg/g PuO ₂ *	± 0.3 µg/g ^(d)
²⁴¹ Am in standard	64.86 mg*	± 0.43 mg ^(e)
Isotopic Weight Fraction		
²³⁸ Pu	0.000149	± 0.000005 ^(f)
²³⁹ Pu	0.929845	± 0.000015 ^(f)
²⁴⁰ Pu	0.067206	± 0.000007 ^(f)
²⁴¹ Pu	0.002141*	± 0.000009 ^(f)
²⁴² Pu	0.000659	± 0.000003 ^(f)
Total Alpha Activity in standard		
	3996 mCi**	± 11 mCi ^(g)
	1.478E+11 Bq	± 4E+08 Bq ^(g)

*Decay corrected value on 6/25/98 ** Effective 6/25/98

Reggie Hunter, Acting
Bill J. McKerley, Group Leader
CMR Analytical Chemistry

Robert S. Marshall
Robert S. Marshall, Project Manager
NDA WRM Production

$$\text{ppm Am } ^{241} = \frac{(64.86 \times 1000) \text{ pg}}{50.01032 \text{ g Pu}} = 1296.93 \pm 8.7$$

Los Alamos National Laboratory Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard Rant 50-3

This NDA standard contains high purity plutonium dioxide dispersed in diatomaceous earth. Quantitative information and uncertainties on the nuclear content of this WRM are listed below. Complete information regarding the Pu and Am content, Pu isotopic ratios, chemical composition, elemental impurities, containment, and fabrication procedures are retained in files by the Los Alamos National Laboratory CMR Analytical Chemistry Group.

NUCLEAR MATERIAL CONTENT

The nuclear material content and total alpha activity for this standard are listed below. Also listed are overall uncertainty estimates at 95% confidence intervals (CI) for each component.

<u>Component</u>	<u>Quantity or Value</u>	<u>95 % CI</u>
PuO ₂ in standard	56.9262 grams	± 0.05298 grams ^(a)
Pu Assay	87.897 % of PuO ₂ *	± 0.081 % ^(b)
Pu in standard	50.03642 grams	± 0.04657 grams ^(c)
²⁴¹ Am Assay	1140.3 µg/g PuO ₂ *	± 0.3 µg/g ^(d)
²⁴¹ Am in standard	64.90 mg*	± 0.43 mg ^(e)
Isotopic Weight Fraction		
²³⁸ Pu	0.000149	± 0.000005 ^(f)
²³⁹ Pu	0.929845	± 0.000015 ^(f)
²⁴⁰ Pu	0.067206	± 0.000007 ^(f)
²⁴¹ Pu	0.002141*	± 0.000009 ^(f)
²⁴² Pu	0.000659	± 0.000003 ^(f)
Total Alpha Activity in standard	3998 mCi** 1.479E+11 Bq	± 11 mCi ^(g) ± 4E+08 Bq ^(g)

*Decay corrected value on 6/25/98 ** Effective 6/25/98

Peggy Bantier, Acting
Bill J. McKerley, Group Leader
CMR Analytical Chemistry

Robert S. Marshall
Robert S. Marshall, Project Manager
NDA WRM Production