Lawrence Livermore National Laboratory
Working Reference Material Production Plan

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*Lawrence Livermore National Laboratory
Working Reference Material Production Plan*

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ABSTRACT

This Lawrence Livermore National Laboratory (LLNL) Working Reference Material Production Plan was written for LLNL by the Los Alamos National Laboratory to address key elements of producing seven Pu-diatomaceous earth NDA Working Reference Materials (WRMs). These WRM's contain low burnup Pu ranging in mass from 0.1 grams to 68 grams. The composite Pu mass of the seven WRM's was designed to approximate the maximum TRU allowable loading of 200 grams Pu.

This document serves two purposes: first, it defines all the operations required to meet the LLNL Statement of Work quality objectives, and second, it provides a record of the production and certification of the WRM's. Guidance provided in ASTM Standard Guide C1128-89 was used to ensure that this Plan addressed all the required elements for producing and certifying Working Reference Materials.

The Production Plan was written to provide a general description of the processes, steps, files, quality control, and certification measures that were taken to produce the WRM's. The Plan identifies the files where detailed procedures, data, quality control, and certification documentation and forms are retained. The Production Plan is organized into three parts: a) an initial section describing the preparation and characterization of the PuO₂ and diatomaceous earth materials, b) middle sections describing the loading, encapsulation, and measurement on the encapsulated WRM's, and c) final sections describing the calculations of the Pu, Am, and alpha activity for the WRM's and the uncertainties associated with these quantities.

I. INTRODUCTION

This Lawrence Livermore National Laboratory (LLNL) Working Reference Material Production Plan addresses the key elements in the production of seven Working Reference Materials (WRMs). These WRM are specified in LLNL Statement of Work (SOW) accompanying Purchase Order #B339558 (Appendix A). These WRM contain PuO_2 blended with diatomaceous earth and were produced using quality objectives, quality control and assurance plans, and procedures developed for producing WRM for the Nondestructive Analysis Performance Demonstration Program (NDA PDP). The seven LLNL WRM addressed in this document contain Pu ranging in mass from 0.1 grams to 68 grams. The composite Pu mass of the seven WRM was designed to approximate the maximum TRU allowable loading of 200 grams Pu.

This document serves two purposes: first, to define all the operations required to meet the SOW objectives as envisioned prior to the start of WRM production, and second, to provide a record of the production and certification of the WRM. Guidance provided in ASTM Standard Guide C1128-89 was used to ensure that this Plan addressed all the subjects appropriate to a production plan.

The Production Plan was written to provide a general description of processes, steps, files, quality control, and certification measures. The Plan identifies the files where detailed procedures, data, quality control and certification documentation, and forms are retained. This Production Plan is organized into three parts: a) an initial section describing the preparation and characterization of the PuO_2 and diatomaceous earth materials, b) middle sections describing the loading, encapsulation, and measurement on the encapsulated WRM, and, c) final sections describing the calculations of the Pu, Am, and alpha activity for the WRM and the uncertainties associated with these quantities.

In appropriate sections throughout this Plan, descriptions are provided of calibrations made using NBS and NIST Reference Materials thereby, providing traceability of critical quantities to Nationally recognized Reference Materials.

Three other documents complement the Production Plan. They are the NDA PDP quality assurance documents: a) the Quality Assurance Project Plan for the PDP II&III WRM Production (PDP II&III 100/0) designating those QA measures implemented to ensure the WRM meet all the quality objectives stated in the SOW, b) the Management Plan (PDP II & III Project Management Plan, PDP II&III-101/0) which defines the management methods and tools used to ensure that the WRM are produced in a cost effective manner while meeting all milestones, and c) the Document Control/Records Management Plan (PDP II&IIIRM Document Control and Records Management Plan, PDP II&III-102/0) that defines those measures taken to ensure that all procedures, records, and data required to document the production, and demonstrate the pedigree, of the WRM are properly generated, documented, retained, and protected until delivered to LLNL.

II. STARTING MATERIALS

A. Plutonium Dioxide

The LLNL WRMs are designed to represent weapons-grade Pu found in TRU waste typically generated on the order of ten to twenty years ago. The PuO₂ selected was purified in 1986 and now contains more than a ten-year daughter product in-growth of ²⁴¹Am. The PuO₂ batch selected (LANL 52/PEOR3257) for the LLNL WRM production was initially prepared by the anion exchange purification of a plutonium nitrate solution followed by precipitation as plutonium oxalate. The resultant Pu oxalate was calcined to PuO₂ and packaged in hermetically sealed food-pack cans. The initial Pu assay, Pu isotopic, and impurity analyses records on this material indicated that it was a suitable starting material, hence the batch was retained for WRM production and was prepared, sampled, and characterized as follows.

1. Preparation

The PuO₂ used for the WRM production was prepared at the Los Alamos National Laboratory (LANL) TA-55 Plutonium Facility. Appendix B contains a detailed report describing all the preparation steps. Sections 1.A - 1.D summarize these steps.

A. Blending and Calcining

The PuO₂ batch was calcined during initial Pu oxalate-to-PuO₂ production and then blended to ensure homogeneity. However, the calcination was not sufficiently rigorous to generate a stable (low surface area) material. Therefore, PuO₂ was placed in a fused silica furnace boat and calcined at 800-950°C for five hours. The furnace temperature history and calcining time was recorded and is detailed in Appendix B.

B. Pulverization

The PuO₂ was a uniform, finely divided powder when initially produced. The PuO₂ was further examined after calcining to determine whether the material homogeneity might be enhanced by grinding. The material appeared finely powdered and highly uniform, and no grinding was required.

C. Screening

The calcined PuO₂ was screened through a 40 mesh sieve. The material passed through the sieve relatively easily indicating that the majority of the particles were ~ 250 μm or less in diameter.

D. Documentation and Records

A record of the calcining time/temperature history, Loss-On-Ignition analysis and particle size analysis were generated and initialed upon completion of the preparation operations. The report is attached as Appendix B and a copy is retained in the LLNL WRM file.

2. Sampling

Quality assurance and quality control (QC) are incorporated into the procedures for all analytical methods used to characterize the PuO₂. The accuracy and precision of these methods have been established via ongoing QC programs and maintained through the routine practice of analyzing control standards. Multiple sampling of the selected PuO₂ batch was required to assess the batch homogeneity and to provide precision estimates for each of the analytical measurements.

A. Sampling Method

A master blend of PuO₂ was prepared and subdivided into four batches containing ~300-500 grams. Material was removed from each of the four batches of PuO₂ and put into a small can, for a total of 95 grams of PuO₂. Next, the 95 g batch of PuO₂ was placed in a container of sufficient diameter to allow removal of five samples from different areas throughout the volume of PuO₂. Samples were removed from different vertical and horizontal regions of the PuO₂ batch using a spatula.

B. Number of Samples

The number of samples selected for the measurement of each attribute was determined from the desired data quality objectives of the NDA PDP WRMs and the demonstrated performance of the instrument/method used to measure the attribute. To insure the desired level of confidence in the data collected from particle size determination, Pu assay, Pu isotopic distribution, Am Assay, and impurity analyses and its subsequent application and propagation, samples of certain size had to be used.

Three samples each were taken for ICP-mass spectroscopy metallic impurities and Ion Chromatography anionic impurities. Two samples were taken for particle size distribution analysis. Four samples were analyzed for Loss on Ignition (moisture content). Five samples were taken for each of the following determinations: Pu Assay, Pu isotopic distribution, and ²⁴¹Am assay (sample dissolution for Pu assay provides aliquots for Pu isotopic distribution and ²⁴¹Am assay). These latter analyses were considered 'critical' analyses because the certified Pu mass, ²⁴¹Am mass, Pu isotopic distribution, and total alpha activity values are used directly to calibrate NDA instrumentation for TRU waste measurements.

C. Packaging and Labeling

All samples were promptly sealed in glass or plastic screw-cap vials after removal from the PuO₂ batch. The vials removed from the sample distribution room were clearly labeled with bar-coded labels with sequential numbers uniquely defining each individual sample. Sample tracking was provided by the LANL Analytical Chemistry Laboratory Information Management System (LIMS).

D. Documentation and Records

Records were made showing sample identification, date, and requested analyses. These records are retained in the LANL LIMS system, and the sample identification and requested analyses are shown in the Appendix C Analytical Chemistry Report. The Analytical Chemistry Report is also retained in the LLNL WRM file.

3. Characterization

A. Particle Size Distribution

Method. A LAB-TEC 1000 Particle Size Analyzer was used to determine the sample particle size distribution. This instrument is a scanning laser microscope that measures particles suspended in a liquid medium. The procedure used for particle size determination is NMT-3-281-MRD-R00. The particle count fractions were determined for particle size distributions with resolution of < 1 micron in 28 increments over the <0.7 to 250 micron range.

Calibration. Quartz standard reference materials BCR No. 67 and 69 were used for particle size analyzer and calibration checks. The instrument laser, detectors, and focal point are fixed in position and cannot be manipulated or adjusted. This very stable instrument was checked using the BCR quartz standard reference materials on a bi-weekly basis. The performance check was performed on the same day as the PuO₂ particle size determination.

Documentation and Reporting The particle size distribution data was reviewed by a project leader prior to using the PuO₂ for preparing WRMs to ensure < 2% of the particles exceeded 150 microns, a NDA PDP quality objective. The Spherical Equivalent Mean diameter of the two samples was 24.2 and 25.6 microns respectively. Less than 0.04% of the particles exceeded 150 microns. Appendix B contains tables showing the detail particle size data. The procedure NMT-3 281-MRD-R00 was used for particle size distribution, LAB-TEC 1000 instrument operation, and calibration. The particle size distribution determined for each sample is retained in the LLNL WRM file.

B. Loss-on-Ignition

Method. Four, five gram samples of the PuO_2 were weighed to the nearest 0.1 milligram into platinum crucibles. The samples were then calcined at 950°C for four hours, cooled in a desiccator, and reweighed. The weight loss was calculated and expressed as percent Loss-on-Ignition.

Calibration. The four place analytical balance used for weighing the samples was calibrated by the Los Alamos Metrology Group. Prior to weighing the balance performance was checked by weighing certified weights.

Documentation and Reporting. The Loss-on-Ignition data was reviewed by the LANL WRM Project Leader prior to using the PuO_2 for preparing WRMs. The average Loss-on-Ignition for the four samples was 0.03 weight percent. An Appendix B data sheet details the weighing data used to calculate the Loss-on-Ignition average value. This Appendix is also contained in the LLNL WRM files.

C. Plutonium Assay

Dissolution. The five sample aliquots taken for Pu assay were independently dissolved. Each sample aliquot was weighed to the nearest 0.01 milligram and the weight recorded. The dissolution method was used ASTM C1168-90 "Standard Practice for Preparation and Dissolution of Plutonium Materials for Analyses." This method uses a sealed reflux tube dissolution, which incorporates a mixture of HCl , HNO_3 , and HF under pressure to ensure complete dissolution.

Solution Preparation. After dissolution, each aliquot was quantitatively transferred to a tared dispensing bottle. Multiple rinses were performed on the dissolution vessel to ensure quantitative transfer. After transfer, the dispensing bottle was weighed to the nearest 0.1 milligram and the net weight of solution recorded. The solution was then thoroughly mixed by swirling and shaking the dispensing bottle for at least 30 seconds.

Standards and Calibration. As described in procedure CST-1-ANC5-035/0, the coulometric titrator was calibrated vs. NBL Certified Reference Material (CRM) #126, providing CRM traceability for sample analysis. All weightings were performed on an analytical balanced certified by the LANL Measurement Technology Group using NIST-traceable weights. Further, titration of the sample aliquots were preceded and followed by titrations of aliquots of a well characterized, high purity PuO_2 control material. The analyses of the control material aliquots were recorded and used as quality control samples to verify the long term precision of the method. The control material assays fell within the 0.5%, three relative standard deviation, coulometric assay control limit. The historical precision of the method is 0.07% at one relative standard deviation. The overall method relative random uncertainty is 0.08%. Inter-laboratory studies have shown that the method has no known bias.

Coulometric Titration. Pu assay values on five samples of the PuO₂ starting material are listed in Table 1. The assays were performed by coulometry as described in procedure CST-1-ANC5-035/0. Two replicate aliquots of each of the dissolved five samples were weight dispensed from the dispensing bottle for coulometric assay. The individual and mean assay values on the five samples can be found in Appendix C.

Table 1. Pu Assay Results, g Pu/g PuO₂

Sample	1	2	3	4	5
g Pu/g PuO ₂	0.8800	0.8789	0.8792	0.8797	0.8782
AVG. g Pu/g PuO ₂ : 0.87915 effective 10/23/96 (decay adjusted to 6/25/98, this value is 0.87897)					

Documentation and Reporting. The sample aliquots weights, dissolution date, net solution weights, coulometric assay results and dates, CRM aliquot assay results, and operator and supervisor initials are recorded in formal LANL notebook ANC49, pp. 80-81. The results of the replicate assays for the five samples are summarized in Table 1 and presented in Appendix C. These data were a technical **HOLD POINT** and were reviewed by the LANL WRM Project Leader prior to preparing the WRM blends.

D. Plutonium Isotopic Distribution and ²⁴¹Am Analysis

The same sample preparation and mass spectrometer operations were used for both Pu isotopic distribution and ²⁴¹Am analysis. The sample preparation and mass spectrometer operation are described in LANL procedure CST-1-ANC5-041/0 "Determination of Uranium and Plutonium Isotopic Distribution and Assay by Mass Spectrometry," CST-1-ANC5-044/0 "Operation of the VG-2 Mass Spectrometer," and CST-1-ANC5-040/0 "Preparation of Pu for Mass Spectrometric Analyses."

Samples. Aliquots of the five solutions prepared for Pu assay (see section II.A.3C) were used for the Pu isotopic distribution and for the ²⁴¹Am assay. Each sample was analyzed in duplicate.

Sample Preparation. For the Pu isotopic distribution determinations, aliquots of the dissolved samples were purified by ion exchange chromatography to separate the Pu and Am fractions and to remove interfering uranium isotopes. The ²⁴¹Am analysis was performed by isotope dilution mass spectrometry and a weight aliquot of each sample was spiked with a known quantity of ²⁴³Am. Using ion exchange chromatography, the Pu fraction and Am fraction were separated and an aliquot of the Am fraction mounted on filaments for the ²⁴¹Am/²⁴³Am mass fraction determination.

Plutonium Isotopic Distribution Analysis. The Pu fraction filaments containing about 20 ng of Pu were mounted in the VG-2 instrument. This instrument is equipped with a multicollector (5) Faraday system, which permits the simultaneous collection of up to five isotopes. The entire filament Pu aliquot is evaporated (Total Evaporation Mass Spectroscopy) and the subsequent ion signal collected thereby yielding a very high precision, high accuracy analysis (see Reference 2). The relative gain of the five measuring channels was determined at the start of each analysis with a built-in current source. The integrated intensity from each isotope was proportional to the abundance of that isotope. The results of these analyses are summarized in Table 2.

Table 2. Summary of Pu Isotopic Distribution as Weight Percent*

Sample	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{244}Pu
1	0.0149	92.9668	6.7198	0.2326	0.0659	0.0000
2	0.0150	92.9653	6.7205	0.2331	0.0661	0.0000
3	0.0155	92.9647	6.7201	0.2337	0.0660	0.0000
4	0.0154	92.9659	6.7210	0.2316	0.0661	0.0000
5	0.0147	92.9678	6.7196	0.2325	0.0655	0.0000
Average	0.0151	92.9661	6.7202	0.2327	0.0659	0.0000

*Effective 10/2/96

^{241}Am Assay. The Am fraction filaments were mounted in the VG-2 Mass Spectrometer and processed as described above. The 241/243 mass ratio was measured and the ^{241}Am concentration in the sample calculated using the 241/243 ratio and the ^{243}Am concentration of the prepared sample. Since the Pu concentration in the sample was known from the Pu assay of the same solution, the $^{241}\text{Am}/\text{Pu}$ mass ratio could be calculated. The $^{241}\text{Am}/\text{Pu}$ assay and $^{241}\text{Am}/\text{g PuO}_2$ results are summarized in Table 3.

Table 3. $^{241}\text{Am}/\text{Pu}$ Analysis Results

Sample	1	2	3	4	5
$\mu\text{g } ^{241}\text{Am}/\text{g PuO}_2$	979.1	979.0	979.5	979.3	979.0
$\mu\text{g } ^{241}\text{Am}/\text{g Pu}$	1113.7	1113.6	1114.1	1113.9	1113.6

Mean $\mu\text{g } ^{241}\text{Am}/\text{g PuO}_2$: 979.2* (1140.3 effective 6/25/98)

One standard deviation: 0.2

*Effective 10/2/96

Standards and Calibration. The Mass Spectrometer is calibrated using NBS SRM-948 aliquots run with every batch of samples receiving Pu isotopic distribution analysis. These data have formed the basis for control charts and were used to assure that the method is meeting performance criteria, and to determine the method's historical performance. Historical QC data demonstrates that this method (sample preparation and instrument performance) is superior to the manufacture's stated precision of 0.05% at one standard deviation.

Documentation and Reporting. The SRM and sample replicate Pu isotopic distribution (both as atomic percent and weight percent) and ^{241}Am assay results, analysis dates, SRM aliquot, and Pu isotopic distribution analysis results (both as atomic percent and weight percent), and operator and supervisor initials are recorded in formal LANL notebook VG2, separation date 10/2/96, with copies introduced into LLNL WRM files. The Pu isotopic distribution results and $^{241}\text{Am}/\text{Pu}$ ratio results for the five samples are summarized in above Tables 2 and 3 and are contained in the "Final Chemical Analysis Report" included as Appendix C. A copy of procedures CST-1-ANC5-040/0, CST-1-ANC5-041/0, and CST-1-ANC5-044/0 are in the NDA PDP PHASE II.A file. The $^{241}\text{Am}/\text{Pu}$ ratio and Pu isotopic distribution results were a technical **HOLD POINT** and reviewed by LANL Project Leader prior to using the PuO_2 to prepare $\text{PuO}_2\text{-DE}$ WRM blends.

E. Inductively-Coupled Plasma Mass Spectrometry Determination of Impurities

The ICP-MS method is well suited for, and was used to determine the concentration of, the following impurities in the PuO_2 : Al, Be, Ca, Fe, Ga, Ni, P, Si, Th, W, Mg, and ^{235}U . The LANL procedure ANC-DE-1-SC-26;R0 was used for sample preparation and the ICP Mass Spectrometric analyses.

Sample Preparation. Three samples of the PuO_2 were dissolved in nitric acid using the LANL microwave dissolution procedure CLS-1-PS-8.01. The dissolved samples were aspirated into an inductively coupled plasma mass spectrometer (ICP-MS) for elemental and isotopic analyses.

ICP-MS Analysis. The samples were atomized and ionized in the plasma, then sampled by a mass spectrometer. Mass detection and concentration measurements were made relative to external calibration for each element. The analysis results for the three samples are summarized in Table 4 below:

Table 4. ICP-MS Analysis Results in Micrograms Impurity Per Gram PuO₂

Impurity	Sample 1	Sample 2	Sample 3	Average
Al	<12.0*	<12*	<12*	<12*
Be	<0.8*	< 0.8*	< 0.8*	< 0.8*
Ca	107	130	130	122
Fe	44	50	50	48
Ga	7.4	7.5	7.7	7.5
Ni	160	100	100	120
P	< 16*	< 16*	< 16*	< 16*
Si	110	100	180	127
Th	1.8	1.0	1.5	1.4
W	338	317	340	332
Mg	< 30*	< 30*	< 30*	< 30*
²³⁵ U	195	209	212	209

* Note: Values preceded by the symbol < represent ICP-MS limits of detection.

Standards and Calibration. Certified Reference Materials traceable to NIST standards for each element were used to calibrate the ICP-MS instrument and to assess instrument performance. The precision and accuracy of this method is +/- 10% based on ongoing calibration and verification standards. Analyses for ²³⁸U was reported but the value in Appendix C is probably that of ²³⁸Pu because ²³⁸Pu is a isobaric interference.

Documentation and Reporting. The sample handling, weights, and dissolution volumes are recorded in formal LANL notebook PQ 2, 101596, and in the LANL CST LIMS system. The complete chemical analysis ICP-MS results for impurity elements are found in Appendix C of this report. Procedure ANC-DE-1-SC-26;R0 was used to analyze the samples by ICP-MS. The sample impurity analysis results were reviewed by the LANL WRM Project Leader to ensure the impurity levels, analysis data, and the batch homogeneity were satisfactory. This review was a technical **HOLD POINT** prior to using the PuO₂ to preparing PuO₂-DE WRM blends.

F. Radiometric Analyses

Radiometric analyses were performed to verify that the ²⁴⁴Cm and ²⁵²Cf levels were low enough to not adversely effect neutron or gamma based measurements of the WRMs and to validate the Mass Spectrometric/IDMS assay of ²⁴¹Am.

Sample Preparation. An aliquot of each of the five PuO₂ samples dissolved for Pu Assay (see section II.A.3.B, "Plutonium Assay") were pipetted onto glass slides and evaporated to dryness for alpha spectra analysis. Also, aliquots from each of the dissolved samples were diluted to 10.0 ml with nitric acid for gamma spectroscopy analysis to determine the ²⁴¹Am concentration.

Radiometric Analyses. The alpha spectra from the five sample aliquots showed no alpha peaks in the energy region 5.7 to 7.0 meV. Based on the alpha background in this energy region and the alpha peak from ²³⁹Pu and ²⁴⁰Pu at 5.155 meV, the measurement established that the ²⁴⁴Cm was <0.06 µg/gPu and the ²⁵²Cf was <0.001 µg/g Pu.

The gamma analysis of the five sample aliquots showed ²⁴¹Am µ/g PuO₂ ratios listed in Table 5:

Table 5. Radiometric Analysis of PuO₂ for ²⁴¹Am

Sample	µg ²⁴¹ Am/g PuO ₂
1	982
2	976
3	977
4	989
5	980

Effective 10/2/98

The mean value of the 5 analyses is 981 µg ²⁴¹Am/g PuO₂. This value is statistically consistent with the more precise Mass Spectrometric value of 979.2 µg ²⁴¹Am/g PuO₂.

Standards and Calibration. The alpha spectrometer was energy calibrated with a source prepared from weapons-grade Pu and ²⁴⁴Cm. This energy calibration was validated vs. NIST Standard Reference Material 4906- C-9, a ²³⁸Pu source.

Documentation and Reporting. The results of the ²⁴⁴Cm and ²⁵²Cf alpha spectrometric analysis and the ²⁴¹Am assay are contained in the Appendix C Analytical Chemistry Report and are recorded in the LANL Analytical Chemistry LIMS system. The alpha spectrometric results for ²⁴⁴Cm and ²⁵²Cf were reported to the LANL WRM project leader and were a technical **HOLD POINT** prior to using the PuO₂ to preparing PuO₂-DE WRM blends.

G. Anionic Impurity Analyses

Chlorine and fluorine potentially add random uncertainty to neutron based waste assays; hence analyses were performed to determine their concentration in the PuO₂. Ion chromatography was used to quantitatively determine chloride and fluoride concentrations.

Sample Preparation. The chlorine and fluorine constituents of three representative samples of PuO₂ were separated from each PuO₂ sample by pyrohydrolysis and collected in a condensate. The pyrohydrolysis procedure is LANL procedure ANC-DE-1-PA-13. Portions of the pyrohydrolyzed condensates were then injected into the ion chromatograph and peaks corresponding to the halides were obtained and compared to peaks obtained from standard solutions.

Ion Chromatography Analysis. The chloride concentrations were determined in the condensates using a Dionex DX-100 ion-chromatograph. The average PuO₂ sample analyses for chloride was 33 micrograms per gram of PuO₂ and for fluoride was 47 micrograms per gram PuO₂.

H. Standards and Calibration

The Dionex-100 instrument was calibrated using a three point calibration curve with VHG solution standards that are traceable to NIST. The instrument was calibrated the day it was used.

I. Documentation and Reporting

The sample aliquots weights, solution volume adjustments and aliquot volumes, sample preparation and analysis dates, CRM assay results, calculations, and operator and supervisor initials are recorded in formal LANL notebook #ANC 89. Procedure CST-STP-ANC5-078/0 was used to analyze samples by Ion Chromatography method. Final data results are recorded in the LANL Analytical Chemistry LIMS system and found in the Appendix C Analytical Chemistry Report. The data was reviewed by the LANL WRM Project Leader and was a technical **HOLD POINT** prior to using the PuO₂ to prepare WRM PuO₂-DE blends.

B. Diatomaceous Earth

J.T. Baker Co. Filteraid diatomaceous earth was obtained in eleven, 1 kg batches. This type of diatomaceous earth was selected instead of the J.T. Baker Co. # 2244-05 (Kieselghur) used for the Phase I WRM production because the Filteraid type was readily available in USA at the time of the study whereas Kieselghur type was not.

1. Preparation

Eleven, 1 kg containers of J. T. Baker "Filteraid" brand diatomaceous earth (DE) were poured into a rigorously cleaned, 55 gallon steel drum and the drum sealed. Using a rotating drum mixer, the DE was blended for 10 hours.

2. Sampling

After blending, the DE was removed in eight increments and placed in eight plastic containers. One sample was taken from each receiving container.

3. Characterization

Characterization of the DE was important to ensure that the material was adequately blended and that the concentration of elements interfering with neutron and gamma based analyses are at or below levels which might introduce significant interference via alpha-n reactions. The constituents of the DE were determined by Glow Discharge Mass Spectrometry (GDMS) using the procedure described by Duckworth, Borshick & Smith, *Journal of Analytical Atomic Spectrometry*, Vol. 8, pp. 875-879, 1993. The GDMS analysis included quantification 29 constituents including the alkaline metals, Be, B, Si, S, Cl, F, Fe, Al, Th, and U.

A. Sample Preparation

Each diatomaceous earth sample (and the SRM materials) was blended with high purity tantalum metal and mounted in the Glow Discharge Mass Spectrometer (GDMS). Eight samples of DE were analyzed by GDMS.

B. Glow-Discharge Mass Spectroscopy Analysis

Glow discharge mass spectrometry is a sputtering technique in which atoms from an electronically conducting sample are introduced into a high-energy argon plasma, ionized, and mass analyzed. The GDMS instrument at LANL is a doubly focusing magnetic sector, high resolution instrument capable of quantification to the nanogram/gram level. The complete report of GDMS data is too extensive to include in text and is contained in Appendix D. A summary of results of constituents with potential neutron interference is provided in Table 7.

Table 7. GDMS Analysis Results on J. T. Baker "Analytical Filteraid" Diatomaceous Earth

Constituent**	Wt %	Constituent	µg/g (ppm)
SiO ₂	87.7	BeO	1.61
Fe ₂ O ₃	1.31	B ₂ O ₃	273
CaO	0.350	Li ₂ O	12.0
Na ₂ O	3.20		
Al ₂ O ₃	4.37		
F	< 0.03 - 0.25		

Table 7 lists the range of F values. The upper limit of F content in DE is < 0.25 weight percent, which is probably biased high. National Institute of Standards and Technology SRM 98b samples were analyzed to measure the precision and accuracy for DE samples by GDMS. There are no certified values reported for F in SRM 98b, hence we cannot unequivocally bias correct our F values.

The constituent values for Na_2O and CaO reported in Table 7 should be considered semiquantitative as the analyses of NIST SRM 98b for these constituents were more than 10% biased.

With the exception of F, the constituents listed in Table 7 and Appendix D are reported as oxides. This reporting is a geologic analysis reporting convention and represents a likely form of the elements in DE but is not necessarily the actual molecular form.

C. Standards and Calibration

The high purity tantalum binder is routinely analyzed for impurities to provide background correction data. NIST SRMs 89, 91, 92, and 98b were combined with the tantalum binder and analyzed to provide calibration sensitivity factors. NIST SRMs 98b was analyzed along with the DE samples as a QC check and to establish method bias.

D. Documentation and Reporting

All pertinent data, including calibration, sample weights, sample identification, sample preparation and analysis dates, QC sample identification, and sample and QC analyses results are recorded on the GDMS hard disk and backup floppy disks of the LANL Inorganic Elemental Analysis Group (CST-8). A copy of the procedure described in Duckworth, Borshick & Smith, *Journal of Analytical Atomic Spectrometry*, Vol. 8, pp. 875-879, 1993 are contained in the LLNL WRM file. A copy of the complete analytical results for all eight samples and the control sample is found in Appendix D. The DE impurity analysis results were reviewed by the LANL WRM Project Leader to ensure the DE homogeneity and impurity content were satisfactory. This review was a technical **HOLD POINT** prior to usage of the DE for preparing WRM blends.

4. Weight Loss

To ensure that the DE did not contain appreciable adsorbed moisture or other volatile species, four samples of the DE were heated to 125°C for more than one hour and the weight loss determined. The weight loss corresponded to an average of 0.11%, indicating that the DE was relatively free of moisture or volatile materials. This measurement data and calculation is found in both Appendix E and the LLNL WRM File. Higher values for weight loss of DE are reported in Appendix D along with GDMS results. However, these results are invalid because of the defective design of the experiment.

III. BLENDING

A Turbula Model T2C blender was used for blending. Separate blends were prepared for each individual WRM to ensure that the amount of Pu in each WRM was known with maximum certainty, e.g. blend homogeneity was not a variable in determining the quantity of Pu in each WRM.

A. Blending Parameters and Blend Evaluations

1. PuO₂-DE Blend Preparation

To evaluate the blending parameters established in cold testing, seven PuO₂-DE blends were prepared corresponding to the seven WRM Pu quantity ranges in the NDA PDP Phase I and IIA WRM Production SOWs. Details of the blend preparation steps are contained in procedure CST-STD-001/1. One hundred twenty four to 135 grams of DE were weighed into 1 liter polyethylene, wide mouth blending bottles. PuO₂ was weighed in an aluminum weighing boat (to the nearest 0.01 mg) and then added to a depression in the center, top surface of the DE in the blending bottle. The weighing boat was weighed after the PuO₂ transfer and the mass of PuO₂ retained in the weighing boat was subtracted from mass PuO₂ weighed out to determine the actual mass of PuO₂ transferred into the blending bottle. The container was sealed, secured in the Turbula blender (Figure 1), and blended for 60 minutes.

2. Homogeneity Evaluation

The procedure used to evaluate blend homogeneity is described in LANL procedure CST-STD-O11/0. Prior to loading of different mass range of diatomaceous earth (DE) and plutonium oxide (PuO₂) blend, the homogeneity of these blends were verified by non-destructive gamma-ray analysis. Seven different mass ranges were evaluated spanning the range 0.03 to 65g of Pu in the blend. The blend homogeneity was determined as follows:

Approximately 20 ml, weighed blend sample was packed into a pre-marked, constant fill height, 30 ml high-precision plastic vial. Six vials of samples were obtained from different portions of the blend. These vials were bagged out from the glovebox with two layers of 4-mil plastic bags. Each sample vial was counted on a high-purity germanium (HPGE) detector at a fixed geometry for each of the seven Pu mass ranges. Each sample was counted and the gamma peak areas determined for the 59.5 keV peak for ²⁴¹Am.

The ²⁴¹Am peak areas were divided by the weight of the sample and the count-per-gram data analyzed to assess the blend uniformity, i.e. consistent mass of ²⁴¹Am per unit mass of the blend. In all cases the standard deviation on the peak area for the six samples was less than the NDA PDP Program homogeneity requirement of less than 2.5% for a 30 milliliter volume of blend.

Blend homogeneity determination results for the NDA PDP Program cannot be supplied as part of this report because the quantity of Pu used in the NDA PDP program WRMs is propriety information. The blend homogeneity data is contained in NDA PDP Phase I and IIA files at LANL and can be produced for auditing purposes as required. The blending data was reviewed by the LANL WRM Project Leader. This review was a technical **HOLD POINT** prior to preparing additional PuO₂-DE blends.

B. WRM Blend Preparation

1. PuO₂ and DE Addition

Prior to producing the first blend, the quantity of DE required to fill a tap-packed stainless steel container to within 0.5 +/-0.05 inches of the top was determined in cold testing to be 135 grams. The amount of DE required for each PuO₂-DE blend to ensure the designed fill height was based on the experience gained from preparing the Phase I and IIA NDA PDP WRMs. The target weight of DE was weighed out to the nearest 0.1 gram in a uniquely identified blend container for each blend and the weight recorded. The containers were introduced into the weighing glovebox, and weighed quantities of PuO₂ were added as described in section III.A.2, "Homogeneity Evaluation." Final weights of the DE and PuO₂ for each WRM blend are listed in Table 8.

TABLE 8. DE and PuO₂ Weighing Data For LLNL Blends

Item	DE, g	g, PuO ₂	as of 6/25/98 amount in Blend	
			g, Pu	μg, 241Am
LLWPU0.1G	133.99	0.11640	0.10231	133
LLWPU1.0G	133.52	1.1358	0.99833	1295
LLWPU10.0G	131.47	11.37627	9.99940	12972
LLWPU25.0G	129.80	28.44401	25.00143	32433
LLWPU65.0G	128.08	73.9536	65.00300	84326
LLWPU67.0G	127.98	76.2028	66.97998	86891
LLWPU68.0G	127.05	77.3546	67.99237	88204

2. Blending

After DE and PuO₂ addition, the blend container lid was secured, and the container was removed from the weighing glovebox, transferred into the blending glovebox, and introduced into the blender. The blending was performed for 60 minutes at the "medium" rotational speed of the Turbula Blender; the conditions established to produce a uniform blend (see section III.A.2, "Homogeneity Evaluation").

3. Documentation and Reporting

Diatomaceous earth weight, PuO₂ weight, blend identification, blend time, date, and operator signature were recorded on the work form from procedure CST-1-STD-001. These forms are retained in the LLNL WRM file.

IV. STAINLESS STEEL ASSEMBLIES

A. Fabrication

The stainless steel assemblies were fabricated from Type 304 stainless steel tubing (seamless) and sheet and bar stock. The inner and outer cylinders and top endcaps were fabricated by a commercial shop following Statement of Work PLA-STD-203A/Rev2 attached as Appendix F. The LANL Main Machine Shop inspected the components, laser cut the bottom endcaps from 304 SS sheet stock, welded these endcaps in place, labeled all the endcaps and helium leak checked the inner cylinder bottom endcap weld. The Statement of Work embracing these activities is PLA-STD-204/2 attached as Appendix F.

The Statements of Work and all QA documentation (manufacture certificates, initial assembly inspection, test component inspection, and leak check records) are contained in the LLNL WRM file.

B. Receiving Inspection

After receipt from the machine shop, the Type 304 stainless steel cylinders and upper endcaps were inspected for cleanliness, identification marks, dimensions (wall thickness, length, diameter), endcap fit, damage, and weld quality. Inspection was performed using LANL procedure CST-STD-006/2, which includes an inspection form.

C. Assembly Identification

The seven assemblies were identified with the following sequential alpha-numeric system: LLWPU0.1G, LLWPU1.0G, LLWPU10.0G, LLWPU25.0G, LLWPU65.0G, LLWPU67.0G, LLWPU68G. The identification was laser-etched into both the assembly inner and outer cylinder endcaps.

D. Documentation

The Statements of Work for the stainless steel assemblies and all QA documentation (manufacture certificates, initial assembly inspection, and leak check records, and final receiving inspections) are contained in the LLNL WRM file.

V. STAINLESS STEEL CYLINDER FILLING

The WRM inner cylinder preparation, filling, packing, frit inserting, bagout for transfer, and cleanup operations are fully described in LANL procedure CST-STD-002/3 and CST-STD-003/2.

A. Cylinder Preparation

To minimize contamination of the inner cylinder, it was wrapped prior to transfer into the glovebox. The cylinder was wrapped with aluminum foil and was taped in a manner to cover all outer exposed areas while leaving the open end exposed for filling. The wrapped cylinder was then labeled to insure that the cylinder was filled from the correct blending

bottle. The cylinder was then placed into the glovebox. The cylinder was fitted with a plastic sleeve that overlapped the lip of the cylinder and protruded down into the tube approximately 0.5 inches. The sleeve prevented contamination of the upper 0.5 inches of the cylinder; the area into which the graphite frit and endcap were placed when filling was completed.

B. Cylinder Filling and Packing

A brown sheet of paper with approximate dimensions of 30X30 cm was placed on the floor of the glovebox. All supplies were kept on top of the brown paper so that if spillage occurred, it could easily be detected. One mg of Diatomaceous Earth (DE) spilled could readily be detected. The cylinders were then placed inside a 4 cm ID graduated cylinder. The graduated cylinder provided stability during the filling and packing of the cylinder. At this stage, a paper funnel was inserted into the cylinder and the blend was added via spatula. A watch glass was placed over the open end of the cylinder to minimize the loss of sample during packing of the material. The material was transferred in small increments and then packed into the container by simultaneously turning the cylinder and tapping it aggressively on the glovebox floor. This was performed until all of the material was transferred into the cylinder. The cylinder was now ready for frit insertion.

C. Frit Insertion

Once all the material was transferred and packed, the paper funnel and plastic sleeve were removed. These actions were done slowly and with care to insure that no material loss occurred. A 0.25 inch thick graphite felt frit was pressed gently into the cylinder on top of the packed material. A plastic bag was then placed over the cylinder to protect the cylinder from contamination from subsequent cylinder loadings.

D. Waste Materials Measurement

The waste materials measurement was the measurement of residual Pu on the items used during the filling procedure. These items include the paper funnel, the tissue used to clean the spatula, plastic sleeve, watch glass and the brown paper used to detect spillage. Each WRM generated a set of waste items and these items were inserted into the WRMs blending bottle and the bottle was bagged out of the filling glovebox and submitted to the radiochemistry team for counting. The measurement was performed using a far-field gamma counting method with a item-to-detector distance of 45 cm. The weight fraction of Pu retained in the waste materials was less than 0.1% in all cases except for WRM LLWPU0.1G which had 0.44 weight percent retention in the waste item. Appendix G describes this measurement method including the calibration methodology and lists the waste item Pu retention for each WRM.

E. Documentation and Records

Records were prepared showing that all the steps pertaining to the cylinder preparation, filling, frit insertion, and cleanup were completed. The records include blending forms showing the quantities of DE and PuO_2 weighed for each WRM and blending times. Cylinder filling forms list cylinder identification numbers, blend identification numbers, and checkoffs for each step required for the cylinder preparation and filling. All records were initialed and dated by the operator performing the tasks.

VII. INERTING

The prepared WRMs were transferred into the inert gas airlock of the welding glovebox. The airlock was evacuated slowly and backfilled with helium gas. This process of evacuating and backfilling was repeated twice. The helium gas served two purposes: one was to prevent oxidation of the stainless steel containers during the welding process and the other was to provide a gas fill for the leak check inspection after the welding operation.

VII. WELDING

A. Welding Setup

A programmable TIG weld power supply was used to perform the TIG welds on both the inner and outer stainless steel tubes.

1. Inner Stainless Steel Tube Welds

The power supply was located outside of the welding glovebox with the welding cables fed into the welding glovebox. Located inside the glovebox was a speed-controlled rotational chuck and a stand holding the welding torch. The stand allowed precise, three directional positioning of the welding electrode. Since the welding was performed in an inert-gas glovebox, no additional inert cover gas was required.

2. Outer Stainless Steel Tube Welds

The outer tube welds were performed outside the glovebox. A speed-controlled rotational chuck and a stand holding the welding torch was used to ensure reproducible welds. An inert cover gas was used to minimize oxidation of the stainless steel during welding.

B. Establishing Weld Parameters

Using the #304 stainless steel cylinders and end caps that comprise the test components (see Appendix F, PLA-STD-203/1), practice welds were performed prior to start of Phase I WRM production. These tests helped establish a welding power supply amperage program, cylinder rotational speed, and welding electrode-to-cylinder distance and position. To ensure acceptable welds, end cap welds were performed at different current levels, and a current operating range was established. These parameters were established for both the inner and outer cylinders and end caps and were also used in the Phase I and Phase II NDA PDP WRM production.

C. Welding Qualification

The welding equipment, welding parameters, welding procedure, and weld operators were qualified as detailed in LANL procedure CST-STD-007/2. Once operational welding parameters were demonstrated through performing test welds, the two welding operators performed qualification welds on both the inner and outer cylinders with appropriate end caps. The welds extended 360 degrees around the cylinders with adequate over-run to assure a complete seal and controlled arc break. The weld qualification acceptance criteria includes visual examination at 10X magnification, helium leak testing, and metallographic examination as defined in LANL procedure CST-STD-005/3. A weld penetration of at least 0.040 inches was demonstrated.

D. Records and Documentation

Dated and signed records were prepared on the weld qualification visual inspection and leak testing. Metallographic examination records include dated and signed photomicrographs of various sections of the qualification weld specimens. These records are retained in the NDA PDP PHASE II.A files and can be produced for audit purposes if requested.

VIII. DECONTAMINATION OF THE INNER CYLINDERS

After the inner cylinder end caps were welded in the inert atmosphere glovebox, the cylinders were removed from the glovebox through a series of cleaning operations. The cleaning was assessed by "smearing" the cylinders with a Health Physics "swipe" cloth and the swipe counted with an alpha swipe counter. Once the cylinders were cleaned to <20 dpm smearable alpha contamination, they were removed from the open-front "glovebox" and placed in plastic bags. The cleaning and contamination measurement procedure was CST-STD-002/3. This procedure and records of the decontamination assessment are retained in the LLNL WRM files.

IX. LEAK CHECKING

A. Helium Leak Check

Helium leak checks were performed on the welder qualification welds for the inner and outer test cylinders and for the inner cylinder welds for every WRM. The helium leak checks were performed following LANL procedure CST-STD-005/3. All cylinders demonstrated a helium leak rate of $< 1 \times 10^{-8}$ atm.-cc/second, meeting Department of Transportation (DOT) requirements for shipping in Class B containment. Dated and initialed leak check records include leak detector calibration data and leak rate data for each WRM.

B. Documentation and Records

The leak check records were delivered to and are retained in the LLNL WRM files. These records were a technical **HOLD POINT** and were reviewed prior to WRM shipment.

X. FINAL INSPECTION

Final WRM inspection included alpha contamination checks, dimensional checks, and gamma-ray measurements to evaluate both WRM Pu uniformity and verify WRM Pu loadings. Each WRM was placed in a plastic bag and sealed after final inspection. The bags were identified using a flow-pen with the same alpha-numeric identifier on the WRM.

A. Alpha Contamination Checks

Thorough alpha contamination measurements were made on every WRM using both "swipe" methods sensitive to 6 dpm/100 cm² removable alpha activity, and direct alpha surveys sensitive to 300 dpm/100 cm². No detectable alpha contamination was found on any WRM. These survey results were recorded, signed, and dated for every WRM and are contained in the LLNL WRM file.

B. Dimensional and Identification Checks

Every WRM was checked for length and diameter to ensure a length of 8.99 to 9.04 inches and a diameter of <1.947 inches. The final weld on the endcap of the outer cylinder did add up to 0.04 inches to the NOMINAL 9.000 inch length of some of the WRMs. Every WRM was visually inspected for any obvious defects such as dents or scratches and to ensure that the identification markings were clear and robust. These data were dated, signed, recorded and retained in the LLNL WRM file.

C. Pu Uniformity Checks

Each WRM was subjected to far field, collimated gamma-ray intensity measurements focused on a 6 cm zone of the left and right (upper and lower if the cylinder is vertically oriented) segments of the cylinder. A high purity germanium detector was positioned 65 cm distant from and orthogonal to the exposed surface of the WRM.

Peak areas for the 59.6 keV gamma peak of ²⁴¹Am and 129, 375, and 413 keV of ²³⁹Pu, were integrated. The most robust data was provided by the 129 keV peak (small counting statistics error and acceptable self-attenuation effects.) The ratio of the left to right end counts were obtained as a measure of uniformity of Pu loading. These ratios are shown in Table 10.

Table 10. Uniformity Measurement Data Summary

WRM ID	Mean L/R Ratio @ 129 keV	Counting Error @ 2 sigma
LLWPU0.1G	0.97	0.11
LLWPU1.0G	1.03	0.055
LLWPU10.0G	0.98	0.033
LLWPU25.0G	1.00	0.023
LLWPU65.0G	1.00	0.023
LLWPU67.0G	1.00	0.024
LLWPU68.0G	1.00	0.023

The left-to-right end count rate ratios confirmed that there is no significant inhomogeneity in the Pu distribution within the WRMs.

Appendix G contains the measurement details and contains a complete compilation of the left-right zone counting data for each WRM. This dated and signed record showing WRM identification and two-zone gamma count data is also contained in the LLNL WRM files.

D. ^{239}Pu Content Verification

The summation of the left and right end counting data provides an assessment of the as-weighed Pu loading in each WRM. The left + right summed peak areas were divided by the weight of PuO_2 for the WRM and the resultant normalized peak areas were plotted. Figure 1 shows the normalized count data and indicates that within counting error, the count areas are consistent with the mass of PuO_2 weighed into each WRM.

Figure 1. Normalized Count Rate for ^{239}Pu 414-keV peak:

Each WRM was sealed in a plastic bag and the bag marked with the WRM identification number plus a "OK" sticker signifying the WRM had passed all inspection and verification steps. The WRMs were wrapped in 'bubble' wrap, inserted into a 2R inner container of a certified DOT 6M shipping containers, and shipped via authorized commercial carriers.

XI. WRM CERTIFICATION

The WRM production process was designed to meet all NDA PDP specified design attributes. To certify the WRMs, critical information and data generated during the WRM production

process was formally documented, filed, and reduced through appropriate calculations to determine the Pu isotope and ^{241}Am mass values and the alpha activity for each WRM. Further, uncertainty data for the critical information and data was propagated to determine the statistical uncertainties of the calculated masses and alpha activities of each completed WRM. Finally, to achieve certification, all critical measurements are traceable to nationally recognized reference materials.

A. Nuclide Mass and Alpha Activity Calculations

The calculation algorithms for Pu mass, Am mass, and alpha activity for the WRMs are presented below. These calculations were executed in the spreadsheet format contained in Appendix H. Also listed in Appendix H are the calculated Pu mass, Am mass, and alpha activities for all seven WRMs.

1. Pu Mass

The net amount of plutonium in each WRM was based on the amount of PuO_2 feed material weighed out for each WRM and calculated as follows:

$$\text{Pu Mass} = (\text{Pu Mass} \times 0.87897^*) - \% \text{ blend loss}/100$$

*effective 10/25/98

% blend loss is the transfer and holdup loss as determined by the gamma spectroscopy assay of the transfer container, the blending container, funnel, cleaning tissues, and the transfer spillage catch paper. For each WRM certificate, the Pu assay value, Pu isotopic composition, and the amounts of Pu, and ^{241}Am were reported as of 6/25/98.

2. ^{241}Am Mass

The ^{241}Am mass for each WRM is calculated as follows:

$$^{241}\text{Am Mass} = (\text{Pu Mass}) \times \text{Am Mass}/\text{Pu Mass}$$

where: **Pu Mass** is that quantity in A.1. above.

Am Mass/Pu Mass is the Am assay result as described in Section II.A.3.E.

3. Alpha Activity

The WRM alpha activity is the summation of the alpha activity contribution from each of the Pu and Am isotopes in the WRM. To facilitate calculating the alpha activity for each WRM, the alpha specific activity (alpha curies per gram PuO_2) for the PuO_2 feed material was calculated effective 6/25/98 as 70.23 mCi/g PuO_2 (79.90 mCi/g Pu). Each WRM alpha activity was calculated by multiplying the WRM PuO_2 mass times 70.23 mCi/g PuO_2 .

Appendix G lists the alpha emitting nuclide half lives used and the alpha specific activity at 6 month intervals from 10/2/96 through 7/1/2002.

B. Uncertainty Data and Calculations

To meet the SOW Section 3.2 and 4.4 criteria for alpha activity and uncertainty, the following information was collected and data reduction performed.

1. Uncertainty Data

A. Plutonium Assay

Pu assay values on five samples of the PuO₂ starting material are listed in Table 1. The assays were performed by coulometry. The individual assay values on the five samples can be found in Appendix C. The mean assay of the five samples was 0.87915 grams Pu/gram PuO₂. The observed standard deviation of the five assays was 0.00064. Since assays from five independent samples were averaged to get the 0.87915 value, the standard deviation of the average was the standard deviation from the coulometry assay values divided by the square root of five. The 95% confidence bound on the assay is therefore $((0.00064/\text{sq. rt. } 5))(2.78) = 0.00080$. Adjusted for ²⁴¹Pu decay, effective 6/25/98 the mean assay value was 0.87987 and the 95% CB was 0.00081g grams Pu/gram PuO₂.

B. Plutonium Isotopic Distribution

The Total Evaporation Mass Spectrometric method used for the Pu isotopic distribution and ²⁴¹Am assay analyses is an absolute method with no known bias. The method was calibrated with the Plutonium Isotopic Standard NBS SRM 948 (Appendix I) which has a Pu isotopic distribution similar to the PuO₂ starting material. The ²³⁹Pu weight percents of the NBS SRM # 948 (normalized to 10/1/96) and the PuO₂ are 91.8444% and 92.9661%, respectively.

The Pu isotopic distribution analyses on the five starting material PuO₂ samples showed standard deviations comparable to those shown Plutonium Isotopic Standard NBS SRM 948, which was measured as a measurement control sample over a four month period immediately prior to the date of the PuO₂ isotopic measurements (Appendix J). To obtain the standard deviations of the average isotopic analyses for the five starting material PuO₂ samples, the observed measurement precision standard deviations were divided by square root of five, the sample size.

These estimates of uncertainties for the stated PuO₂ isotope weight percents are multiplied by a t-distribution value of 2.78 to provide 95% confidence bounds (CB) and are listed in Table 11. The Table 11 data has been decay adjusted to 6/25/98.

Table 11. Error Estimates for Mass Spectrometric Pu Isotopic Distribution

Isotope	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu
Mean*	0.0149	92.9845	6.7206	0.2141	0.0659
Std. Dev.	0.00037	0.00122	0.00056	0.00078	0.00025
95% C.B.	0.00046	0.00152	0.00070	0.00087	0.00031

* The isotope ratio values are decay corrected to 6/25/98

C. Americium Assay

The ^{241}Am assay values for the five samples of PuO_2 analyzed are listed in Table 3. The mean assay value of 979.2 micrograms $^{241}\text{Am}/\text{g PuO}_2$ has a one standard deviation uncertainty of 0.22 micrograms $^{241}\text{Am}/\text{g PuO}_2$. Since assays from five independent samples were averaged to get the 979.2 micrograms $^{241}\text{Am}/\text{g PuO}_2$ value, the standard deviation of the average was the standard deviation from the five analyses divided by the square root of five. The 95% confidence bound on the average assay is 0.27 micrograms $^{241}\text{Am}/\text{g PuO}_2$. The 95% confidence bound expressed as relative percent is 0.028%.

The isotope dilution mass spectrometric method uses a ^{243}Am spike as an internal standard. The relative uncertainty of the ^{243}Am solution concentration has been estimated as 0.25% at 95% confidence. This estimate was made based on the original solution assay (EDTA titration) and comparative studies on EURATOM samples (PIDIE) [Reference 3]. The 0.25% random uncertainty serves as a systematic uncertainty, as applied to the ^{241}Am assay; hence, it is conservatively added directly to the random uncertainty 95% confidence bound for the five sample assays. Therefore the total relative percent uncertainty for the ^{241}Am assay has an estimated 95% confidence bound of $\pm 0.28\%$, relative.

D. Weighing Uncertainty

Random uncertainties for weighing were estimated from repeated weightings and were performed using a standard weight most closely matching that of the target quantity of PuO_2 weighted. These weightings were performed before each set of same-target-mass PuO_2 was weighed. The balance performance is summarized in Table 12 below and full control weighing data is listed in Appendix K.

Table 12. Precision on Standard-Control Weight Weightings

Wt range	AVG.	1 SD	at 95% CL	
			SD	% RSD
20 mg	0.019998	5.00E-06	0.000016	0.0800
100 mg	0.100015	0.000008	0.000022	0.0220
5 g	5.000037	0.000011	0.000027	0.0005
10 g	10.00005	0.000079	0.000019	0.0002
20 g	20.00002	0.000021	0.000045	0.0002
50 g	50.00016	0.000075	0.00016	0.0003
100 g	100.0002	0.000139	0.00032	0.0003

The standard weight set used for control weightings was certified against NIST traceable weights. A certification for the weight set and analytical balance is contained in Appendix L. The observed control weight checks (Table 12) show no statistically significant bias from the certified mass for the weights. Hence the WRM PuO_2 weights were considered free of significant bias.

E. Plutonium Content Left in the Waste Item

Transfer and holdup loss quantities were measured by counting the 59.6 keV peak of the ^{241}Am contained in the funnels, cleaning tissues, and spillage catch paper used to transfer the blend material into the stainless steel cylinder. The percent of blend loss to the waste item was calculated based on the initial amount of ^{241}Am in the blend (as of 5/1/97) and the amount of ^{241}Am left in the waste item (as of 5/1/97). The percent of blending material retention was very low ($< 0.1\%$) except for low-mass WRM LLWPU0.1G with 0.44% retention in the waste. The percent of ^{241}Am left in the waste items is listed in Table VI of Appendix G. Since the ^{241}Am is the decay product of ^{241}Pu , it was assumed that the percent of Pu in the waste items was identical.

Uncertainty terms for these waste item measurements include the preparation of the ^{241}Am standard, the gamma spectroscopy instrumentation calibration, and the uncertainty associated with the counting of each waste item. The dominating uncertainty term (three relative percent at one standard deviation) associated with the counting of each item is associated with the positioning of the item and was determined in a positional/rotational study for the NDA PDP PHASE IIA WRM production.

The uncertainty associated with the gamma spectroscopy counter calibration includes uncertainty in the standard value used for calibration, uncertainty in the calibration curve shape, and uncertainty in the calibration short term variability. From prior studies on pipette calibration, weighing, and the calibration curve fit, the calibration short term random variability was estimated to be small. The relative effect of the uncertainty in the calibration curve shape and the standard value depends on count rate of the waste item and was assumed to be a systematic error.

The WRM waste item uncertainty terms are listed in detail in Appendix G, and each WRM's waste item Pu content and uncertainty component is incorporated in the Appendix H tables.

F. Alpha Activity

Alpha half-life values for converting Pu and Am isotope quantities in the WRMs to alpha activity were presented on certificates for the NBS SRM 946 Plutonium Metal and NIST SRM 4322B Alpha Particle Solution Standard (^{241}Am) (see Appendix I). The certificate for SRM 946 did not include uncertainty estimates for the listed Pu isotope half-lives, so values were obtained from the literature (References 4 and 5). These uncertainty values, and those from the SRM 4322B Certificate for ^{241}Am , are listed in Table 13.

Table 13. Uncertainty Values for Isotope Half-lives

Isotope	Half-life, Years	Half-life Uncertainty,* Years
²³⁸ Pu	87.84	0.04
²³⁹ Pu	24119	26
²⁴⁰ Pu	6560	11
²⁴¹ Pu	14.34	0.02
²⁴² Pu	387,000	900
²⁴¹ Am	432.2	0.5

* Half life uncertainties for ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu (Reference 4); for ²⁴⁰Pu, ²⁴¹Pu (Reference 5), and for ²⁴¹Am (NIST SRM 4322B Certificate).

The alpha specific activity for the feed PuO₂, discussed in section XI.A.3, has three uncertainty components: alpha half-life, Pu isotopic abundance, and Am isotopic abundance.

These data are listed in Table 13 (half-life uncertainty), Table 11 (Pu isotopic ratio uncertainties), and Section B.1.C. (²⁴¹Am isotopic abundance uncertainty). The alpha specific activity and its associated uncertainty for the feed PuO₂ uses the above data and the error propagation methodology described in the Appendix M document "Validation of Plutonium Oxide PEOR3258-STD-1, As The Primary Radiochemistry Standard." The alpha activity and calculated 95% confidence limit for each WRM is listed in the tables in Appendix I.

G. Combined Uncertainties For Mass And Alpha Activity

Standard uncertainty estimation methods were used to calculate individual random and systematic uncertainties for Pu, PuO₂, Am mass, alpha activity, and Pu isotope ratios. Propagation of variance (POV) techniques were used to appropriately combine the uncertainties for all the components used to calculate nuclear quantities. Where well-supported estimates on uncertainties for components were not available, a conservative approach was taken to use values that are believed to exceed the actual values. Therefore, the 95% confidence intervals (bounds) presented in this Plan, and listed on the WRM Certificates, are believed to be larger than they would be if exhaustive data were available for every uncertainty component.

Uncertainty components included in estimates of the 95% confidence interval for the five nuclear quantities listed on the WRM Certificates are identified in Table 14.

Table 14. Components Included in Uncertainty Calculations

Nuclear Quantity/ Uncer. Source	Weighing	Pu Assay	Pu Isotope Ratio	Am Transfer	Counting
PuO ₂ Mass	X				
Pu Mass	X	X		X	X
Am Mass	X	X		X	X
Pu Isotopic			X		
Alpha Activity	X	X	X	X	X

Appendix H lists the numeric values for each step of the uncertainty calculation. These steps provide the estimate the 95% confidence limits for the quantities listed in Table 14. Appendix N Certification of Content and Traceability documents summarize the critical parameters (Pu mass, Am mass, and alpha activity) and their associated uncertainties for all the LLNL WRMs.

C. WRM CERTIFICATION

The WRMs are formally certified by a combination of multiple analyses characterizing the PuO₂ and DE starting materials, the calibration and check of measurement methods with Certified or Standard Reference Materials, the Project Quality Assurance Plans, the numerous signed and dated QA forms used to document inspections and completion of fabrication steps, the review of calculations and critical documents, the statistical analysis of data using standard data reduction methods, the qualification and training of persons responsible for analysis and fabrication operations, and the formal control and filing of documents.

To provide a single document attesting to the formal certification of the seven WRMs, Certificates of Content and Traceability for each WRM were prepared, signed, and dated by the LANL CMR Analytical Chemistry Group Leader, Bill J. McKerley, and the LANL Project Leader for LANLWRM production, Robert S. Marshall. A copy of each WRM Certificates is included as Appendix N.

The reduction of PuO₂ characterization, mass transfer data, and control data generated during the production of the WRMs produces the Pu isotope mass and alpha activity and uncertainty data required for the LLNL WRM certification. These data and calculations are formally recorded, signed and stored in the LLNL WRM file. Certification also requires that the critical characterization measurements (Pu assay, Pu isotopic distribution, Am assay, and PuO₂ weighings) be traceable to nationally recognized reference materials. This traceability has been demonstrated by identifying these reference materials in this Production Plan and in the analytical procedures used. The original Certificates were delivered to the LLNL customer, Kem Hainebach. Copies of these reference material certificates are retained in the LLNL file

and included in this document as Appendix N. To the highest degree possible, traceability to nationally recognized reference materials was established for the less critical parameters including particle size analysis, impurity analyses, isotope half-life and alpha specific activity, and helium leak check.

ACKNOWLEDGEMENTS

Over 50 talented and generous persons contributed to the many and varied tasks required to produce the LLNL Pu WRMs. To name all these persons is impossible! We are very grateful for the TA-55 persons under the guidance of Tim Hayes for preparing and packaging the PuO₂ source materials and performing the particle size and LOI analyses. Our thanks for the Yeamans machine shop and LANL ESA Machine shop for the fine work they performed machining, inspecting, welding, labeling, and leak checking the stainless steel components. Let us single out Vince Hesch for guiding these components through the multiple ESA Machine Shop tasks and performing the beautiful laser welding and labeling! Our thanks to our many analytical chemistry group colleagues for performing the multiple Pu assay, Am assay, Pu isotopic, radiochemistry, and halide and metallic impurity analyses. The quality of these characterizations are central to our NDA reference materials projects. Equally critical to the quality of our WRM projects are two QA persons we would like to recognize for their most professional, efficient and courteous assistance: Marilyn Foley and Michele Gubernatis. Don Vance deserves special thanks for his development of the final helium leak check and amazingly effective (over 6 orders of magnitude!) decontamination methods. Finally we would like to thank our welding engineer Dave Shapland for the welding system design, and we extend special thanks to Toby Romero for performing all the final welds with skill and professionalism, as well as adding humor to our days!

Many thanks to Kem Hainebach, our customer. Kem provided that level of technical guidance, patience during our unavoidable delays, and keen interest that made this project especially gratifying. We hope these WRMs provide excellent service to Kem and his LLNL colleagues.

REFERENCES

1. NDA PDP Working Reference Materials Production Plan - Phase I, CST-8-PLA-STD-104, LAUR-96-2277.
2. E.L. Callis and R.M. Abernathy, *International Journal of Mass Spec. and Ion Processes*, 103 (1991), pp93-105).
3. Chauvenet and J. Morel, "Intercomparaison des mesures de Composition Isotopique du Plutoniuim par Spectrometrie X et Gamma Resultats de L'action 'Pieie,' Raport Final," report CEA-R-5582.
4. W. Strohm, *International Journal of Applied Radiation Isotopes*, 29, 481-483 (1979).
5. W. Strohm, *International Journal of Applied Radiation Isotopes*, 35, 155-157 (1984).

A

UNCLASSIFIED -- Facsimile Cover Sheet

**Los Alamos National Laboratory
P.O. Box 1663, MS P292
Los Alamos, NM 87545**

To: Bob Marshall**Fax #:** (505) 665-4737**Date:** August 27, 1997

Confirmation #: (505) 667-0409

From: Barbara Carmichael **Fax #:** (505) 665-4853
BUS-2, MS P292

Confirmation #: (505) 667-0264

No. of Pages (Including Cover Sheet): 4

Bob - attached is Mod #2 from Livermore on your order. Please review the scope of work for acceptance.

In order to set up a program code, I will need a UBET estimate from Pat Medina for the funding amount, and an ES&H form. Currently the period of performance on this order is 9/30/97.

If you have questions, please contact me. Thanks.

Barbara

AUG-27-97 WED 10:25 AM LLNL PROGRAM SUPPORT DIV FAX NO. 510 422 8588

P. 02/04

University of California
Lawrence Livermore National Laboratory
Procurement and Materiel DOE Contract W-7405-ENG-48)

August 26, 1997

Ms. Barbara Carmichael, MS P292
Los Alamos National Laboratory
P.O. Box 1663
Los Alamos, New Mexico 87544

Subject: Modification No. 02 to Integrated Contractor Order (ICO) B339558

Enclosed is a copy of the subject ICO. The Los Alamos National Laboratory Representative for this ICO is Bob Marshall. Please return a signed copy of the ICO to me via fax or mail. My mailing address is P.O. Box 808, Mail Code L-650 Livermore, CA. 94550. If you have any questions, you can contact me by phone at (510) 422-0559 or by FAX on (510) 423-7643.

Sincerely,

Linda Denyer
ICO Subcontract Administrator
Procurement and Materiel

Enclosures: as noted

Modification No. 02 to

Integrated Contractor Order (ICO)
B339558

between

The Regents of the University of California

and

Los Alamos National Laboratory

INTRODUCTION

This Modification No. 02 to ICO B339558 is entered into by and between The Regents of the University of California, and Los Alamos National Laboratory.

PURPOSE

The purpose of this Modification No. 02 is to change the Scope of Work and add funding to this ICO.

FUNDING

From: [REDACTED]

By: [REDACTED]

To: [REDACTED]

AGREEMENT

Delete Article II in its entirety and replace with the following:

"II. ESTIMATED COSTS AND LIMITATIONS

The estimated cost of the work to be performed is [REDACTED]. The University has currently obligated [REDACTED] as the maximum amount to be reimbursed to Los Alamos National Laboratory. Los Alamos National Laboratory shall not incur costs or make commitments in excess of this amount without prior written approval of the University Subcontract Administrator.

Los Alamos National Laboratory shall maintain cost information and notify the LLNL Subcontract Administrator named herein whenever it becomes apparent that the cost of performance of any ICO will exceed the amount obligated by the University."

Scope of Work

Delete Attachment A Scope of Work in its entirety and replace with the following:

"Los Alamos National Laboratory (LANL) shall fabricate five (5) sources in sealed containers identical to the NDA PDP working reference materials except that the amounts of radioactive material in each are as follows:

Source 1 shall have 0.1 gm of weapons grade plutonium dispersed in diatomaceous earth, labeled: LL W Pu 0.1g

Source 2 shall have 1.0 gm of weapons grade plutonium dispersed in diatomaceous earth, labeled: LL W Pu 1.0g

B339558

Modification No. 02

AUG-27-97 WED 10:26 AM

LLNL PROGRAM SUPPORT DIV

FAX NO. 510 422 8598

P. 04/04

Source 3 shall have 10.0 gm of weapons grade plutonium dispersed in diatomaceous earth, labeled: LL W Pu 10.0g

Source 4 shall have 25.0 gm of weapons grade plutonium dispersed in diatomaceous earth, labeled: LL W Pu 25.0g

Source 5 shall have 65.0 gm of weapons grade plutonium dispersed in diatomaceous earth, labeled: LL W Pu 65.0g

Source 6 shall have 67.0 gm of weapons grade plutonium dispersed in diatomaceous earth, labeled: LL W Pu 67.0g

Source 7 shall have 68.0 gm of weapons grade plutonium dispersed in diatomaceous earth, labeled: LL W Pu 68.0g

Include Q documentation for each source."

All other terms, conditions and provisions of the ICC shall remain in full force and effect.

Effective upon Los Alamos National Laboratory's signature acceptance of this Modification No. 02, ICC B339558 is hereby amended by mutual agreement of the parties.

ACCEPTED:

Los Alamos National Laboratory

BY: _____

TITLE: _____

DATE: _____

AUTHORIZED:The Regents of the
University of CaliforniaBY: • Linda DenyerLinda Denyer
TITLE: Subcontract Administrator

Lawrence Livermore National Laboratory

DATE: 8-27-97

B339558

Modification No. 02

Integrated Contractor Order
B339558

between

The Regents of the University of California

and

Los Alamos National Laboratory

I. INTRODUCTION

The Regents of the University of California, hereinafter called "University", have entered into Contract No. W-7405 ENG-48 with the United States Government, hereinafter called "Government", represented by the Department of Energy, hereinafter called "DOE", for the management and operation of the Lawrence Livermore National Laboratory, hereinafter called "LLNL". This Integrated Contractor Order (ICO) is in support of the work provided for under the said prime contract.

This document is not a contract. It is an ICO issued for services, equipment, or materials as specified in the "Scope of Work" in Attachment A to be supplied under Government Contract No. W-7405-ENG-36 between the U.S. Department of Energy and Los Alamos National Laboratory which are required in connection with Government Contract No. W-7405 ENG-48 between the DOE and the University.

II. ESTIMATED COSTS AND LIMITATIONS

The estimated cost of the work to be performed is \$16,385.99. The University has currently obligated **\$16,385.99** as the maximum amount to be reimbursed to Los Alamos National Laboratory. Los Alamos National Laboratory shall not incur costs or make commitments in excess of this amount without prior written approval of the University Subcontract Administrator.

Los Alamos National Laboratory shall maintain cost information and notify the LLNL Subcontract Administrator named herein whenever it becomes apparent that the cost of performance of any ICO will exceed the amount obligated by the University.

III. ACCOUNTING FOR COSTS

Accounting for the cost of the services, equipment, materials, and travel shall be made in accordance with and as required by the DOE "Accounting Practices and Procedures Handbook as revised", and Government contract No. W-7405-ENG-36.

IV. PERIOD OF PERFORMANCE

This ICO shall commence upon acceptance by Los Alamos National Laboratory and shall continue through **September 30, 1997**. Los Alamos National Laboratory shall not be reimbursed for costs incurred which are not within this period of performance. Any further periods of performance shall be the result of mutual agreement of the parties and a Modification shall be issued in writing by the University.

V. INVOICES

Billings by Los Alamos National Laboratory shall be monthly and be sent in duplicate to:

University of California
Accounts Payable
P.O. Box 808, L-432
Livermore, CA 94551
Attention: ICO No. B339558

Invoices shall be prepared in accordance with Los Alamos National Laboratory standard, DOE approved invoice format. Invoices must contain the information requested by the "Property" and "Travel" Articles of this ICO. Interest shall not be paid on delinquent payments. Los Alamos National Laboratory shall provide Laboratory with an invoice marked "Final Invoice," notifying Laboratory that no further invoices and charges are forthcoming. In addition, Los Alamos National Laboratory shall provide Property Disposition Information as requested from the Laboratory during ICO Closeout at the conclusion of the term of the ICO. Laboratory will immediately process final invoices and make every effort to pay final invoices within 30 days of (i) acceptance by LLNL of all required reports or other deliverables, and (ii) upon the conclusion of the Property Disposition Process.

VI. PROPERTY

The following property is authorized under this agreement:

Los Alamos National Laboratory Acquired:
none

LLNL Furnished:
none

When acquiring property with University funds, Los Alamos National Laboratory shall include a Property Identification List (Attachment B) with the invoice, requesting payment. Title to all property acquired under this ICO vests in DOE/LLNL.

VII. TRAVEL

Travel is authorized as follows:

Domestic Travel:

<u>Traveler(s)</u>	<u>Destination/Purpose</u>	<u>Date(s) or Frequency</u>
none		

Foreign Travel:

<u>Traveler(s)</u>	<u>Destination/Purpose</u>	<u>Date(s) or Frequency</u>
none		

Invoices seeking reimbursement for travel, must contain an itinerary detailing the same information as above.

VIII. SHIPPING AND DELIVERY INSTRUCTIONS

Ship: Best Way Collect

University of California
Lawrence Livermore National Laboratory
for U.S. Department of Energy
CO: B339558
7000 East Avenue
Livermore, CA. 94550
For: Material Management, Bill Ruvalcaba, Bldg 231, VAULT
Delivery hours-8:00 a.m-4:45 p.m.only

MARKING OF PACKAGES:

All deliverable items shall include on the exterior of the shipping carton the ICO number and name and telephone number of the LLNL Technical Representative. All shipments shall include a packing list.

This package shall be marked HAZARDOUS MATERIAL - RADIOACTIVE

IX. COORDINATION AND ADMINISTRATION

The University's Technical Representative is **Kem Hainebach** at L-546, phone 510-422-4572, who shall represent the University in matters relating to the technical performance of the Scope of Work described herein. The University's Technical Representative shall interpret the technical requirements of the Scope of Work and determine the emphasis and direction of Los Alamos National Laboratory in the conduct of the work.

The University's Subcontract Administrator is **Linda Denyer** at L-650, phone 510-422-0559. All matters relating to the non-technical interpretation, administration, and performance of this ICO shall be reserved to the University's Subcontract Administrator. Los Alamos National Laboratory shall direct all notices and requests for approval to the University's Subcontract Administrator and any notices or approvals from the University to Los Alamos National Laboratory shall be issued by the University's Subcontract Administrator.

All Modifications to the Scope of Work or to the "Estimated Costs and Limitations" Article must be in writing and accepted by both the University and Los Alamos National Laboratory.

X. TERMINATION

The University has the unilateral right to terminate this ICO by written notice to Los Alamos National Laboratory. The University shall reimburse Los Alamos National Laboratory for all cost incurred and all non-canceled costs, up to the amount in the Article entitled "Estimated Costs and Limitations".

XI. ATTACHMENTS

This ICO consists of this document along with the following attachments:

1. Attachment A - Scope of Work
2. Attachment B - Property Identification List

ACCEPTED:

Los Alamos National Laboratory

BY: _____

TITLE: _____

DATE: _____

AUTHORIZED:

The Regents of the
University of California

BY: *Linda Denyer*
Linda Denyer

TITLE: Subcontract Administrator
LAWRENCE LIVERMORE NATIONAL
LABORATORY

DATE: 5-20-97

ATTACHMENT A

SCOPE OF WORK

Los Alamos National Laboratory (LANL) shall fabricate three (3) sources in sealed containers identical to the NDA PDP working reference materials except that the amounts of radioactive material in each are as follows:

Source 1 shall have 0.1 gm of weapons grade plutonium dispersed in diatomaceous earth.

Source 2 shall have 1.0 gm of weapons grade plutonium dispersed in diatomaceous earth.

Source 3 shall have 10.0 gm of weapons grade plutonium dispersed in diatomaceous earth.

ATTACHMENT B
PROPERTY IDENTIFICATION LIST

Vendor: Los Alamos National Laboratory

ICO No.: B339558

Invoice No.:

DOE/LLNL Property No.	Property/Equipment Description & Model	Serial No.	Date Acq.	Location	Qty.	Total Cost

Capital Equipment: Purchases of \$5,000.00 or more

Low Value Equipment: Purchases from \$1,000.00 to \$4,999.99

Attractive Equipment: Purchases of \$300.00 or more including these selected items:
 Electronic Balances, Binoculars, Cellular Phones, Personal Computers (incl. Portables & Laptop), Telescopes, VCR-Type Video Cameras, Still Cameras (excludes closeup, powder, x-ray, oscilloscope, Polaroid, photomacrographic, photomicrographic), Video Tape Recorders & Players, Radio Pagers (no minimum purchase cost), Computer Workstations, Night Vision Devices, TV Camera/Video Disk, Firearms (no minimum purchase cost), Facsimile Machines, TV Receivers/VCR Unit, Laser Style & Color Printers, Portable Video Projector, Two-Way Radio (no minimum purchase cost), Color Video Printer, Video Walkman.

B

Los Alamos

NATIONAL LABORATORY

memorandum

*Nuclear Materials Processing -
Actinide Process Chemistry*
NMT-2

To/MS: R. S. Marshal, CST-8, G740
Thru: J. D. Williams, NMT-2, Group Leader
From/MS: T. A. Hayes, NMT-2, E511
Phone/FAX: 5-8267/ FAX 5-1780
Symbol: NMT-2:FY97-008
Date: October 11, 1996

SUBJECT Plutonium Oxide Treatment and Characterization Data

Several weeks ago I sent you plutonium oxide items PEOR3257A, PEOR3257B, PEOR3257C, PEOR3257D, and a oxide sample PEOR3257S. This memorandum contains data relevant to these oxides.

The original item PEOR3257 was selected based on the available chemistry results (first attachment). The item was calcined in dry air using the following furnace program:

From (*C)	To (*C)	Ramp/Soak time (Hours)
Room Temp	200	1
200	400	1
400	600	1
600	800	1
800	950	2
950	950	2
950	700	2
700	500	2
Furnace	off	

During the initial furnace cycle the material was observed to reach 950 *C. However, during the night the furnace chart recorder failed so there was no document confirming the furnace run. A second run was done and the chart is attached (second attachment). It is quite possible the material went through two furnace cycles. The original item before calcination weighed 1464.3 grams; after calcination the item weighed 1455.7 grams (a loss of 8.6 grams).

The item was mixed and split into 5 items, the four I sent to you and a larger item sent to Gary Rinehart. Three (3) samples were taken from these items, one for LOI (third attachment), another for particle size analysis (forth attachment), and the third was sent to you (PEOR3257S). These samples were taken by removing equal cuts from each of the 5 items.

If you have any questions or further clarification is needed on the processing of these items, please contact me.

att/as

FIRST ATTACHMENT: ORIGINAL ANALYTICAL DATA FOR PEOR3257

Analytical Chemistry Report

Sample No.: 63563

Date Reported: 22-AUG-88

Submitting Group: MST-12

Submitter: HAPKE

Form B Code: K530

Sample Identification: PEORS3257

Date Submitted: 08-AUG-88

Sample Description: PU OXIDE

Analyses Requested: Pu , PuIso, Th , U , Am , SpecA, Rpt

ANALYTICAL RESULTSAnalytical Results

Pu* = 87.1%

DJT

Analyst(s) Notebook Page(s)

EL

324

144

(*) % = g/100 g Sample and ppm = ug/g Sample

COMMENTS: *Sample was weighed in an inert box.

Analytical Chemistry Report

Sample No.: 63563

Date Reported: 25-AUG-88

Submitting Group: MST-12

Submitter: HAPKE

Form B Code: K530

Sample Identification: PEORS3257

Date Submitted: 08-AUG-88

Sample Description: PU OXIDE

Analyses Requested: Pu , Pulso, Th , U , Am , SpecA, Rpt

ANALYTICAL RESULTSAnalytical Results

Wt% Pu-238 = 0.016
Wt% Pu-239 = 93.1
Wt% Pu-240 = 6.53
Wt% Pu-241 = 0.343

Analyst(s) Notebook Page(s)

LM

RC

P.D.S.

(*) % = g/100 g Sample and ppm = ug/g Sample

COMMENTS:

Analytical Chemistry Report

Sample No.: 63563

Date Reported: 31-AUG-88

Submitting Group: MST-12

Submitter: HAPKE

Form B Code: K530

Sample Identification: PEORS3257

Date Submitted: 08-AUG-88

Sample Description: PU OXIDE

Analyses Requested: Pu , PuIso, Th , U , Am , SpecA, Rpt

ANALYTICAL RESULTSAnalytical ResultsAnalyst(s)Notebook Page(s)

1 Th = 20 ppm

BJ

4078

74

TKM

8 8 7 0

(+) % = g/100 g Sample and ppm = ug/g Sample

COMMENTS:

Analytical Chemistry Report

Sample No.: 63563

Date Reported: 29-AUG-88

Submitting Group: MST-12

Submitter: HAPKE

Form B Code: K530

Sample Identification: PEORS3257

Date Submitted: 08-AUG-88

Sample Description: PU OXIDE

Analyses Requested: Pu , Pu Iso, Th , U , Am , SpecA, Spi

ANALYTICAL RESULTS

Analytical Results

Analyst(s) Notebook Page(s)

U = 25 ppm

JMH, MPM

A 5493 111

JMH

COMMENTS:

(P) % = g/100 g Sample and ppm = ug/g Sample

Analytical Chemistry Report

Sample No.: 63563

Date Reported: 24-AUG-88

Submitting Group: MST-12

Submitter: HAPKE

Form B Code: K530

Sample Identification: PEORS3257

Date Submitted: 08-AUG-88

Sample Description: PU OXIDE

Analyses Requested: Pu ,Puiso ,Th ,U ,Am ,SpecA ,Rpt ,Rpt

ANALYTICAL RESULTSAnalytical ResultsAnalyst(s) Notebook Page(s)

Am = 12.6 ppm
+/- 0.43 %rsd
on 8-23-88

LM,MAA

RC

.
..
.

7105.

(*) % = G/100 G Sample and PPM = uG/G Sample

COMMENTS:

Analytical Chemistry Report

Sample No.: 63563

Date Reported: 1-SEP-88

Submitting Group: MST-12

Submitter: HAPKE

Form B Code: K530

Sample Identification: PEORS3257

Date Submitted: 08-AUG-88

Sample Description: PU OXIDE

Analyses Requested: PU , PuIso, Th , U , Am , SpecA, Rpt

ANALYTICAL RESULTS

Aq 30	Al <5	B <5	Be <1	Bi 5	Ca 750-
Cd <10	Cr 200	Cu 5	Fe 140	Mg 50	7500
Mn 2	Mo 300-	Na 50	Ni 390	Pb 15	Si 140
Sn <5	3000	Zn <5			

Our standards are based on trace impurities in pure PuO₂. Experience has shown that if any one impurity level exceeds 2000 ug/g or the summation of impurity levels for several impurities exceeds 3000 ug/g, results for other impurities may be biased.

PEC
WMMW

COMMENTS: Plate No. 498382

Spectro Qual: Result reported to an order of magnitude concentration range.
Spectro Quant: Result reported by a single number generally has a factor of two accuracy with a precision of 50% relative standard deviation. If the number is followed by an *, the precision is between 5 and 20% RSD.

Inductively Coupled Plasma (ICP): Result reported by a single number, followed by an *, has an accuracy within 20% relative of the true value and a precision of 3 to 5% RSD.

Other precisions, if attained, are indicated after the number. Result is based on sample as received and is expressed as ug/g or ug/ml unless indicated otherwise. Result preceded by a < symbol represents our present limit of detection.

QA Procedure:

SECOND ATTACHMENT: FURNACE STRIP CHART RECORD

92817

42817

CHART NO. B956ASS

YOKOJIMA

THIRD ATTACHMENT: LOSS ON IGNITION DATA

DETERMINATION OF LOSS-ON-IGNITION OF PLUTONIUM DIOXIDE OR OTHER
PLUTONIUM-CONTAINING POWDERS - DATA RECORD

SAMPLE ID PEOR32576J DATE 9/11/96
FURNACE CALIBRATION NO. 638783
CALIBRATION EXPIRATION DATE 8/8/97
IGNITION TEMPERATURE 950 °C IGNITION TIME 4 HRS.

ALIQUOT #1

ALIQUOT #2

CRUCIBLE NO. 57CRUCIBLE NO. 58SAMPLE WEIGHT & TARE

BEFORE IGNITION (B1T1) 32.5490 g. (B2T2) 32.5919 g.
TARE (T1) 27.5373 g. (T2) 27.2835 g.
INITIAL SAMPLE WEIGHT (B1) 5.0117 g. (B2) 5.3084 g.

SAMPLE WEIGHT & TARE

AFTER IGNITION (A1T1) 32.5488 g. (A2T2) 32.5908¹¹⁰⁰ g.
TARE (T1) 27.5373 g. (T2) 27.2835 g.
FINAL SAMPLE WEIGHT (A1) 5.0115 g. (A2) 5.3076 g.

PERCENT LOSS (L1) 0.0039 % (L2) 0.0150 %AVERAGE LOSS ON IGNITION .00945 % ALIQUOT SPREAD .0111 %OPERATOR SIGNATURE Charles Davis DATE 9/12/96CHECKED BY CARL MARTINEZ DATE 9/12/96

DETERMINATION OF LOSS-ON-IGNITION OF PLUTONIUM DIOXIDE OR OTHER
PLUTONIUM-CONTAINING POWDERS - DATA RECORD

SAMPLE ID PEOR3257LOI DATE 9/17/96
FURNACE CALIBRATION NO. 638783
CALIBRATION EXPIRATION DATE 8/8/97
IGNITION TEMPERATURE 1000 °C IGNITION TIME 1 HRS.

ALIQUOT #1

ALIQUOT #2

CRUCIBLE NO. 57CRUCIBLE NO. 58SAMPLE WEIGHT & TARE

BEFORE IGNITION (B1T1) 32.5500 g. (B2T2) 32.5921 g.
TARE (T1) 27.5373 g. (T2) 27.2835 g.
INITIAL SAMPLE WEIGHT (B1) 5.0127 g. (B2) 5.3086 g.

SAMPLE WEIGHT & TARE

AFTER IGNITION (A1T1) 32.5477 g. (A2T2) 32.5898 g.
TARE (T1) 27.5373 g. (T2) 27.2835 g.
FINAL SAMPLE WEIGHT (A1) 5.0104 g. (A2) 5.3063 g.

PERCENT LOSS (L1) 0.0459 % (L2) 0.0433 %

AVERAGE LOSS ON IGNITION 0.0446 % ALIQUOT SPREAD 0.0026 %

OPERATOR SIGNATURE Charles J. Davis DATE 9/18/96

CHECKED BY _____ DATE _____

FORTH ATTACHMENT: PARTICLE SIZE DATA

LAB-TEC PARTICLE ANALYSIS SUMMARY

Ver. 5.20.07

HAYES PEOR3257PS

Sample ID : CUT #1
CF Code :
Date : 01/03/80
Time : 01:58:18
Spherical Equivalent Mean : 24.2
Standard Deviation : 23.8
Skewness : 0.3
Kurtosis : 7.9
Coeff. of Variance(%) : 98.4
Counts : 16136

Cycles : 20
Scaling : 1400

M.Cycle Time: 1.0 sec.
Averaged last 5 cycle(s)

Channel Size (microns)	%/Channel	Channel Size (microns)	%/Channel
<0.7	5.5556	25	7.4711
0.7	1.2152	31	6.6649
1.0	1.5824	37	6.2535
1.3	1.1174	44	5.6716
1.8	1.1755	53	4.3210
2.3	2.2640	63	2.9209
3.0	3.3074	75	1.9178
4.0	3.1895	88	1.0326
5.0	4.3516	105	0.5827
6.5	6.1415	125	0.3072
8	7.3506	149	0.1162
10	6.3980	177	0.0295
13	5.7192	210	0.0054
17	6.2153	250	0.0031
20	7.1192

LAB-TEC PARTICLE ANALYSIS SUMMARY

Ver. 5.20.07

HAYES PEOR3257PS

Sample ID : CUT #1
CF Code :
Date : 01/03/80
Time : 01:51:22
Scanned Counts Mean : 26.5
Standard Deviation : 35.1
Skewness : 0.4
Kurtosis : 12.2
Coeff. of Variance(%) : 132.7
Counts : 15575

Cycles : 20
Scaling : 1.00

M.Cycle Time: 1.0 sec.
Averaged last 5 cycle(s)

Channel Size (microns)	%/Channel	Channel Size (microns)	%/Channel
<0.7	11.0301	25	5.2157
0.7	2.2182	31	5.0049
1.0	2.5228	37	4.7965
1.3	2.3331	44	4.3827
1.8	2.4284	53	3.7133
2.3	3.1789	63	3.1790
3.0	3.6708	75	2.5466
4.0	3.8045	88	2.0121
5.0	4.2698	105	1.3955
6.5	4.8274	125	1.0683
8	5.0737	149	0.6870
10	4.9267	177	0.3958
13	4.7750	210	0.2201
17	5.0168	250	0.1552
20	5.1511

LAB-TEC PARTICLE ANALYSIS SUMMARY

Ver. 5.20.07

HAYES PEOR3257PS

Sample ID : CUT #2
CF Code :
Date : 01/03/80
Time : 02:16:24
Spherical Equivalent Mean : 25.6
Standard Deviation : 25.5
Skewness : 0.3
Kurtosis : 7.4
Coeff. of Variance(%) : 99.6
Counts : 15392

Cycles : 20
Scaling : 1:00

M.Cycle Time: 1.0 sec.
Averaged last 5 cycle(s)

Channel Size (microns)	%/Channel	Channel Size (microns)	%/Channel
<0.7	5.6875	25	7.6037
0.7	0.9567	31	6.4428
1.0	1.3203	37	5.6853
1.3	1.0624	44	5.4674
1.8	1.0537	53	4.5698
2.3	1.9040	63	3.4348
3.0	2.8303	75	2.2771
4.0	3.2729	88	1.5013
5.0	4.3860	105	0.8253
6.5	5.7551	125	0.4485
8	7.1336	149	0.1560
10	6.9976	177	0.0231
13	5.9772	210	0.0103
17	6.0729	250	0.0031
20	7.1413

LAB-TEC PARTICLE ANALYSIS SUMMARY

Ver. 5.20.07

HAYES PEOR3257PS

Sample ID : CUT #2
CF Code :
Date : 01/03/80
Time : 02:11:38
Scanned Counts Mean : 28.2
Standard Deviation : 36.9
Skewness : 0.4
Kurtosis : 11.2
Coeff. of Variance(%) : 130.9
Counts : 14819

Cycles : 20
Scaling : 1:00

M.Cycle Time: 1.0 sec.
Averaged last 5 cycle(s)

Channel Size (microns)	%/Channel	Channel Size (microns)	%/Channel
<0.7	10.7357	25	5.2053
0.7	2.0797	31	4.8615
1.0	2.3592	37	4.6266
1.3	2.2427	44	4.5856
1.8	2.2518	53	4.0677
2.3	3.0575	63	3.4803
3.0	3.5665	75	2.7653
4.0	3.7746	88	2.1839
5.0	4.0490	105	1.7002
6.5	4.7526	125	1.2555
8	5.0912	149	0.8326
10	5.0165	177	0.4138
13	4.6397	210	0.2518
17	4.8355	250	0.1911
20	5.1265		

C

Analytical Chemistry Report

Sample ID: 200022136 Group: CST-8 QA: Y Priority: 2
Submission ID: 100017550 Acct: Y Logged: 25-Sep-1996
User sample ID: PEOR3257S
Sample Type: AH PLUTONIUM OXIDE
Customer Cost Code: M592
Requester: B. MARSHALL MS: G740 Phone: 7-4087

Sample Description: PU OXIDE
Radioactive Elements: PU239
Radiation: Y
Hazards: N
Special Handling: N
Final Report Date: 14-Jul-1997

Approved by: System Manager, SQL*LIMS 11-Dec-1996 21:05

Analytical Operation: PACPU-S Task ID: 300068143
Date Completed: 23-Oct-1996 10:31 Replicates: 5 Version: 1

Component	Rep 1	Rep 2	Rep 3	Rep 4	Rep 5	Units
PLUTONIUM	88.00 [1]	87.89 [2]	87.92 [3]	87.97 [4]	87.82 [5]	percent

Assay Method: Coulometry

Estimated Precision: 0.1%

[1] PLUTONIUM, Rep 1:

Average of duplicate analysis. 88.01% and 87.94%

[2] PLUTONIUM, Rep 2:

Average of duplicate analysis. 87.89% and 87.89%

[3] PLUTONIUM, Rep 3:

Average of duplicate analysis. 87.90% and 87.94%

[4] PLUTONIUM, Rep 4:

Average of duplicate analysis. 88.00% and 87.94%

[5] PLUTONIUM, Rep 5:

Average of duplicate analysis. 87.81% and 87.83%

Notebook references: Rep 1: ANC49 PP.80-81

Rep 2: SAME

Rep 3: SAME

Rep 4: SAME

Rep 5: SAME

Analysts: Rep 1: WJB,EL

Rep 2: SAME

Rep 3: SAME

Rep 4: SAME

Rep 5: SAME

Approved by: Temer, Donald J. 23-Oct-1996 10:33

Analytical Operation: MSPUIISO

Task ID: 300068144

Date Completed: 29-Oct-1996 08:57 Replicates: 5

Version: 1

Component	Rep 1	Rep 2	Rep 3	Rep 4	Rep 5	Units
PU-238	0.0150	0.0151	0.0156	0.0155	0.0147	atom percent
PU-239	92.9955	92.9939	92.9933	92.9947	92.9964	atom percent
PU-240	6.6938	6.6945	6.6941	6.6950	6.6936	atom percent
PU-241	0.2307	0.2312	0.2318	0.2297	0.2306	atom percent
PU-242	0.0651	0.0653	0.0652	0.0653	0.0647	atom percent
PU-244	0.0000	0.0000	0.0000	0.0000	0.0000	atom percent
SUM	100.0001	100.0000	100.0000	100.0002	100.0000	atom percent
PU-238	0.0149	0.0150	0.0155	0.0154	0.0146	weight percent
PU-239	92.9668	92.9653	92.9647	92.9659	92.9678	weight percent
PU-240	6.7198	6.7205	6.7201	6.7210	6.7196	weight percent
PU-241	0.2326	0.2331	0.2337	0.2316	0.2325	weight percent
PU-242	0.0659	0.0661	0.0660	0.0661	0.0655	weight percent
PU-244	0.0000	0.0000	0.0000	0.0000	0.0000	weight percent
COMMENTS	[1]	[2]	[3]	[4]	[5]	

Component	Average	Units
PU-238	0.0152	atom percent
PU-239	92.9948	atom percent
PU-240	6.6942	atom percent
PU-241	0.2308	atom percent
PU-242	0.0651	atom percent
PU-244	0.0000	atom percent
SUM	100.0001	atom percent
PU-238	0.0151	weight percent
PU-239	92.9661	weight percent
PU-240	6.7202	weight percent
PU-241	0.2327	weight percent
PU-242	0.0659	weight percent
PU-244	0.0000	weight percent
COMMENTS	n/a	

[1] COMMENTS, Rep 1: 238PU BY RADIOCHEM/MS

[2] COMMENTS, Rep 2: 238PU BY RADIOCHEM/MS

[3] COMMENTS, Rep 3: 238PU BY RADIOCHEM/MS

[4] COMMENTS, Rep 4: 238PU BY RADIOCHEM/MS

[5] COMMENTS, Rep 5: 238PU BY RADIOCHEM/MS

Notebook reference: VG2, SEPARATION DATE 10/2/96

Analyst: NMS,CH,JA

Approved by: Callis, Larry 29-Oct-1996 09:35

Analytical Operation: RCAM

Date Completed: 02-Oct-1996 08:47 Replicates: 1

Task ID: 300068145

Version: 1

Component	Result	Units
-----	-----	-----
AMERICIUM-241	[1]	ppm
%RSD +/-	0.55	
COMMENTS	[2]	

[1] AMERICIUM-241: 982,976,977,989,980 ON 9/30/96

[2] COMMENTS: DUPLICATE ANALYSIS RAN ON 5 CUTS

Analyst: Townsend, Lisa

Approved by: Stalnaker, Nelson D. 02-Oct-1996 08:49

Analytical Operation: PSMPU-S

Date Completed: 06-Dec-1996 00:00 Replicates: 1

Task ID: 300068146

Version: 1

Component	Results	Units
-----	-----	-----
ALUMINUM	<12	ug/g
BERYLLIUM	<0.8	ug/g
CALCIUM	107	ug/g
IRON	44	ug/g
GALLIUM	7.42±0.02	ug/g
NICKEL	160	ug/g

PHOSPHORUS	<16	ug/g
SILICON	110	ug/g
THORIUM	1.80±0.00	ug/g

TUNGSTEN	338.6	0.3	ug/g
COMMENTS	CUT 22		
MAGNESIUM	<30		ug/g
URANIUM 235	195	38	ug/g
URANIUM 238	131	25	ug/g

Notebook reference: PQ2 101596

Analyst: CAB

Approved by: Yoshida, Thomas M 11-Dec-1996 08:43

Analytical Operation: IC-ANIONS-S

Task ID: 300068147

Date Completed: 04-Oct-1996 14:52 Replicates: 1

Version: 1

Component	Result	Units
FLUORIDE	47	ug/g
CHLORIDE	33	ug/g

Notebook reference: ANC 89

Analyst: VMM

Approved by: Sweetin, Deborah 04-Oct-1996 14:55

Analytical Operation: RCESS

Task ID: 300068148

Date Completed: 24-Oct-1996 10:40 Replicates: 1

Version: 1

Component	Result	Units
SURVEY	[1]	
COMMENTS	[2]	

[1] SURVEY: PU238 VALUES-3.39,3.41,3.52,3.49,3.32% %RSD=2.34

ALPHA SPECTROSCOPY WAS PERFORMED ON CST#22136. 5 CUTS OF SAMPLE WERE ANALYZED FOR 244CM AND 252CF. NO EVIDENCE OF HIGH ENERGY ALPHA PARTICLES <5.7-7.0 MEV> WAS FOUND. THE ALPHA SPECTRA ARE KEPT WITH THE SAMPLE ANALYSIS SHEET. THE ESTIMATED DETECTION LIMIT FOR 244CM IS 0.06 PPM, FOR 252CF IS 0.001 PPM.

[2] COMMENTS: SAMPLE SPECTRA ARE KEPT IN THE FILE.

Analyst: WONG, STALNAKER

Approved by: Stalnaker, Nelson D. 24-Oct-1996 10:43

Analytical Operation: TED1

Date Completed: 21-Oct-1996 09:32 Replicates: 1

Task ID: 300068149

Version: 1

Component	Result	Units
DISSOLUTION	Y	Y/N

Notebook reference: 10733, P. 128

Analyst: TRH

Approved by: Marshall, Thomas K. 21-Oct-1996 09:34

Analytical Operation: MSAM-S

Date Completed: 29-Oct-1996 09:30 Replicates: 5

Task ID: 300068151

Version: 1

Component	Rep 1	Rep 2	Rep 3	Rep 4	Rep 5	Units
AMERICIUM	979.1	979.0	979.5	979.3	979.0	ug/g

Notebook reference: VG2, SEPARATION DATE 10/2/96

Analyst: NMS, CH, JA

Approved by: Callis, Larry 29-Oct-1996 09:35

Analytical Operation: PSMPU-S

Date Completed: 06-Dec-1996 00:00 Replicates: 1

Task ID: 300071187

Version: 1

Component	Results	Units
ALUMINUM	<12	ug/g
BERYLLIUM	<0.8	ug/g
CALCIUM	130	ug/g
IRON	50	ug/g
GALLIUM	7.5 $\pm$ 0.4	ug/g
NICKEL	100	ug/g
PHOSPHORUS	<16	ug/g
SILICON	100	ug/g
THORIUM	1.0 $\pm$ 1.1	ug/g
TUNGSTEN	317 $\pm$ 25	ug/g
COMMENTS	CUT 23	
MAGNESIUM	<30	ug/g
URANIUM 235	209 $\pm$ 9	ug/g
URANIUM 238	140 $\pm$ 6	ug/g

Notebook reference: PQ2 101596

Analyst: CAB

Approved by: Yoshida, Thomas M 11-Dec-1996 08:43

Analytical Operation: PSMPU-S

Task ID: 300071189

Date Completed: 06-Dec-1996 00:00 Replicates: 1

Version: 1

Component	Results	Units
ALUMINUM	<12	ug/g
BERYLLIUM	<0.8	ug/g
CALCIUM	130	ug/g
IRON	50	ug/g
GALLIUM	7.7 $\pm$ 0.5	ug/g
NICKEL	100	ug/g
PHOSPHORUS	<16	ug/g
SILICON	180	ug/g
THORIUM	1.5 $\pm$ 1.3	ug/g
TUNGSTEN	340 $\pm$ 20	ug/g
COMMENTS	CUT 24	
MAGNESIUM	<30	ug/g
URANIUM 235	212.1 $\pm$ 0.5	ug/g
URANIUM 238	142.1 $\pm$ 0.3	ug/g

Notebook reference: PQ2 101596

Analyst: CAB

Approved by: Yoshida, Thomas M 11-Dec-1996 08:43

D

DIATOMACEOUS EARTH SAMPLES (page 1)

	T-3		T-2		T-2		B-2		B-8	
	AVG	STD	Wt% Oxide	(1-SIGMA)	AVG	STD	Wt% Oxide	(1-SIGMA)	AVG	STD
Na2O	2.99	0.25	2.64	0.31	4.14	0.08	3.08	0.04		
MgO	0.795	0.007	0.871	0.023	0.822	0.012	0.756	0.017		
Al2O3	4.22	0.11	4.18	0.25	4.67	0.05	4.33	0.10		
SiO2	87.2	1.1	88.4	1.2	86.2	1.0	87.8	1.0		
P2O5	0.147	0.005	0.145	0.011	0.136	0.007	0.125	0.003		
SO3	0.063	0.007	0.065	0.003	0.073	0.004	0.064	0.004		
K2O	0.408	0.008	0.331	0.066	0.505	0.011	0.337	0.031		
CaO	0.327	0.031	0.392	0.015	0.346	0.008	0.290	0.023		
TiO2	0.225	0.009	0.215	0.012	0.217	0.004	0.224	0.013		
Fe2O3	1.33	0.02	1.30	0.02	1.20	0.01	1.20	0.04		
C	0.055	0.007	0.181	0.015	0.048	0.001	0.076	0.002		
F	< 0.25		< 0.025		< 0.12		< 0.03			
% LOI	1.95		1.171		1.469		1.573			
Selected Trace elements										
Li2O	10.0	2.2	11.2	2.7	12.9	1.0	10.8	1.6		
BeO	1.47	0.06	1.68	0.17	1.61	0.09	1.60	0.23		
B2O3	266	2	308	10	262	4	244	10		
MnO	60	8	187	7	65	9	68	9		
ZrO2	203	38	206	20	118	13	174	20		
BaO	195	8	206	20	183	9	213	7		
PbO2	3.49	0.17	6.35	0.36	5.87	0.56	5.26	0.96		

Analyst: David M. Wayne, CST-8
Method: secondary cathode Glow Discharge Mass Spectrometry

DIATOMACEOUS EARTH SAMPLES (page 2)

	B-4		B-3		T-8		T-4	
	AVG	STD	AVG	STD	AVG	STD	AVG	STD
	Wt% Oxide	(1-SIGMA)	Wt% Oxide	(1-SIGMA)	Wt% Oxide	(1-SIGMA)	Wt% Oxide	(1-SIGMA)
Na2O	3.44	0.21	3.52	0.17	3.10	0.15	2.72	0.12
MgO	0.824	0.017	0.809	0.037	0.791	0.021	0.862	0.032
Al2O3	4.47	0.10	4.52	0.23	4.07	0.11	4.48	0.20
SiO2	87.7	2.9	87.7	3.0	88.1	0.9	88.3	1.9
P2O5	0.161	0.005	0.133	0.003	0.132	0.004	0.134	0.003
SO3	0.067	0.004	0.063	0.004	0.066	0.003	0.057	0.004
K2O	0.408	0.029	0.465	0.030	0.362	0.011	0.324	0.025
CaO	0.359	0.023	0.364	0.014	0.324	0.011	0.387	0.031
TiO2	0.226	0.007	0.218	0.002	0.213	0.016	0.237	0.015
Fe2O3	1.23	0.02	1.19	0.09	1.74	0.02	1.31	0.01
C	0.034	0.004	0.029	0.000	0.042	0.006	0.038	0.002
F	<0.06		<0.03		<0.035		<0.037	
% LOI	0.874		0.874		0.979		0.985	

Selected Trace elements

	ppm	(1-SIGMA)	ppm	(1-SIGMA)	ppm	(1-SIGMA)
Li2O	14.8	1.3	14.1	1.5	8.6	0.7
BeO	1.62	0.12	1.60	0.10	1.44	0.18
B2O3	303	4	286	2	252	8
MnO	64	5	55	5	46	3
ZrO2	183	7	126	9	368	84
BaO	212	7	207	6	206	3
PbO2	7.52	0.17	6.89	0.49	7.37	0.83

Analyst: David M. Wayne, CST-8

Method: secondary cathode Glow Discharge Mass Spectrometry

E

1/25/97 DE LOSS-ON-IGNITION

To assess possibility samples of DE have elevated moisture as indicated by the GDMS analysis report, I performed the following:

- dried 4 beakers by heating 1 hr @ 140°C, cooled in desiccator
- was put on balance & tared to zero
- Poured in sample, recorded weight
- Placed in oven @ 125°C
- Removed after 1 hr, cooled in desiccator
- weighed dried sample.

Date:	Sample Container ID	1B	5B	6B	7B
Beaker #	1	2	3	4	
Tared Beaker?	yes	yes	yes	yes	
WT sample	1.658	1.878	1.784	1.725	
Time w oven*	12:19	12:19	12:19	12:19	
Time out oven	2:20	2:20	2:20	2:20	
Final wt(gms):	53.257	36.886	25.571	34.122	
Final Tare =	51.600	29.010	23.791	32.398	
net wt	1.657	1.876	1.780	1.724	
% wt change	-0.001	-0.002	-0.004	-0.001	
	0.06%	0.11%	0.22%	0.06%	

* Oven temp varied from 100°C to 150°C during drying (cycling)

Ave wt loss = 0.11%
 σ = 0.075%
 95% OCL = 0.23%

DAN 1/25/97

Above weighings performed on Sartorius balance
 PN 839672, QA File # 13045. Calib. expiration date 3-31-97

F

NDA PDP WRM STATEMENT OF WORK SEAMLESS STAINLESS STEEL TUBING AND ENDCAP FABRICATION

8/18/98

LANL/NMT-1 Document # PLA-STD-203A/rev2

Summary: The work requested is : a) cutting to length and machining of 100 inner and outer stainless steel tubes , b) the fabrication of top endcaps for the above stainless steel tubes and, c) fabrication of an additional 50 top endcaps for the stainless steel tubes.. A critical specification is that the close-fitting inner tubes must freely slide inside the outer tubes.

The materials specifications, machined tubing and endcap dimensional specifications, quantities, delivery dates and quality assurance details are as follows:

Material Specifications

1. Tubing: The SS tubing shall be seamless drawn with a chemical composition of 304 stainless steel (SAE type 30404 or AISI type 304).
2. Bar stock for endcaps shall be 304 stainless steel (SAE type 30404 or AISI type 304).

These material shall be ordered and supplied by the Los Alamos National Laboratory.

Item Dimensions

1. Two tube sizes are required with the following specifications (see Figure 1 illustrations):
 - a) Outer Tube Dimensions
 - O.D. - $1.940 + 0.000", - 0.007"$
 - Wall - $0.065 \pm 0.005"$
 - height - $9.000 - 0.000, + 0.005"$
 - b) Inner Tube Dimensions
 - O.D.- $1.800 + 0.000", - 0.005"$
 - wall - $0.045 \pm 0.005"$
 - height $8.685 + 0.000, - 0.005"$

2. One top endcap for each of the inner and outer tubes, is required (bottom endcaps shall be fabricated by Los Alamos National Laboratory). The top endcaps shall be machined from 304 SS flat or bar stock. The inner and outer tube endcap configuration and dimensions are as specified in Figure 2. An illustration detail of the top inner and outer top endcaps in the assembled configuration is also provided for clarity, see Figure 3.

a. Outer Tube Top Endcap Dimensions

(Refer to Figure 2)

O.D., - 1.81 " and machined to provide 0.003" clearance fit to outer tube

Thickness, - 0.210 " +/- 0.002"

Groove, - See Figure 3. Bottom radius of groove not critical.

b. Inner Tube Top Endcap Dimensions

(Refer to Figure 2)

O.D., - 1.71 " and machined to provide 0.0005 " interference fit to outer tube

Thickness, - 0.201 " +/- 0.005"

Groove, - See Figure 2. Bottom radius of groove not critical.

Taper, - A 5° lead angle taper shall be provided on the face opposite from the groove face. See Figure 2.

Machined Part Cleaning/Fabrication

The inner and outer SS seamless tube and associated endcaps are to be machined to specification with burrs and sharp edges removed. Note that the inside concentricity of the as delivered tubing has in the past varied a few thousands of an inch. To prevent the machined tubing wall thickness from falling below the required specification, it is advised that the Outer Tube tubing be machined to an average OD closer to the maximum OD (1.940") rather than the minimum OD (1.933").

All component external surface finish shall correspond to 63RMS or better. Inside surface finish for the tubes may be as-delivered (or equivalent) by the tubing manufacture.

All components shall be delivered free of residual oil or other contaminants. Cleaning with a detergent or solvent is acceptable.

Quantity

Each inner tube, outer tube, inner tube top endcap, and outer tube top endcap constitute an assembly. One hundred such assemblies are required which are comprised of the following components:

- 100 inner seamless tubes,
- 100 inner tube endcaps,
- 100 outer seamless tubes,
- 100 outer endcaps.

An **additional** 25 inner tube endcaps and 25 outer tube endcaps are required.

Inspection

The components shall be inspected during the course of fabrication to ensure all dimensional and finish specifications are met. Records of such inspection are not required. Of particular importance is a) the un-obstructed insertion of the inner tube into the outer tube, b) the clearance fit of the outer tube endcap and c) the light interference fit of the inner tube end cap. The Los Alamos National Laboratory shall perform 100% quality assurance inspection on all components and shall only accept components meeting the specifications listed in this document.

Packaging

The tubing and endcaps shall be packaged in sealed plastic bags such that they are isolated from contaminants. If inner tubes and endcaps are fabricated to fit specific tubes (matched components), these components shall be packaged together and separate from other components. The packages of matched components shall be uniquely identified with sequential numbers (permanent flow-pen lettering on the plastic bag is acceptable).

Delivery Time

The date the Los Alamos National Laboratory awards this job and delivers the tubing and bar stock materials shall be termed the *start date*. The first set of 25 completed assemblies shall be delivered to the NMT-1 Project Manager within 14 working days after the *start date*. The second set of 75 completed assemblies shall be delivered to the NMT-1 Project Manager within 60 working days after the *start date*.

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Figure 1. Stainless Steel Double Encapsulation

Outer Cylinder Top End Cap (Style A)

Inner Cylinder Top End Cap (Style B)

* Approximate wall thickness. Machine to match actual tube wall thickness.

Figure 2. Top Inner and Outer End Cap Dimensions

Revision to
Fig 2, FLA-STD-103/1

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Figure 3 Ton Inner and Outer End Cap Assembly Detail

STATEMENT OF WORK SEAMLESS STAINLESS STEEL TUBING ASSEMBLIES

8/18/98

LANL/NMT-1 Document # PLA-STD-204/2

Page 1 of 6

Introduction

Seamless drawn stainless steel (SS) tubing has been specified to serve as the encapsulation material for the fabrication of a set of radioactive standards. The standards are comprised of two cylinders each, one fitting tightly within the other. The inner cylinder, with bottom endcap TIG or laser welded in place, is filled with a substrate/radioactive material mixture. Once dispensed, the top inner cylinder endcap is TIG welded in place. The inner cylinder is then smoothed to outside diameter and height specifications removing projections inadvertently introduced during the welding process, if required. The completed inner cylinder is inserted into the outer cylinder, bottom endcap previously TIG or laser welded in place, and the outer cylinder top endcap is TIG welded in place followed by deburring. This constitutes the completed unit, a sealed radioactive material/substrate doubly encapsulated in seamless SS tubes with welded endcaps.

The critical requirement associated with the fabrication of the SS encapsulation cylinders is that the radial and axial clearance between them be such that there is no perceptible movement of the inner cylinder relative to the outer in the completed assembly. This dictates that careful consideration be given to machining tolerances and the subsequent fitting of the six components to the assembly; inner tube, outer tube, two inner endcaps, and two outer endcaps. Therefore the dimensions and tolerances specified below are to be interpreted as enveloping values where minor adjustments to the various components may be required to achieve the stated interior and exterior dimensions and subsequent fitup. The double cylinder SS encapsulation device configuration is illustrated with dimensions in Figures 1 through 4, attached.

This Statement of Work describes the work requested of the Los Alamos National ESA/WMA Laboratory Main Machine Shop. Associated with this plan is a similar SOW written to have a local Los Alamos commercial machine shop prepare lengths of tubing and endcaps. Briefly, ESA/WMA will order the required materials, deliver tubing and endcap materials to the commercial machine shop, inspect the commercial machine shop product, prepare and weld bottom endcaps, laser label all endcaps, helium leak check all welded tube/endcap assemblies, perform final inspection and packaging, and provide all material certification, helium leak check and dimensional inspection documentation.

The following are the material, fabrication and testing requirements for the cylindrical SS encapsulation devices. Following are Quality Assurance requirements for the assemblies and delivery details.

Material

1) The SS tubing shall be seamless drawn with a chemical composition of 304 stainless steel (SAE type 30404 or AISI type 304). Appropriate tubing will be ordered by the ESA/WMA Main Machine Shop and delivered to the designated commercial machine shop.

2) Two tube sizes are required with the following specifications (see Figure 1 & 2 illustrations):

a) Outer Tube Dimensions

O.D. -	1.940 + 0.000", - 0.007"
Wall -	0.065 +/- 0.005"
height -	9.000 - 0.000", + 0.005"

These Outer Tubes will be fabricated by the designated commercial machine shop.

To facilitate fitment of the bottom endcap, a pedistal fixture will be used to locate the endcap flush with the tube end. This endcap will be fabricated and welded in place by the ESA/WMA Main Machine Shop. The top endcap will be located by insertion into the outer tube until it bottoms out on the top of the inner tube. The top endcap will be welded in place by NMT-11 at the CMR Building.

b) Inner Tube Dimensions

O.D.-	1.800 + 0.000", - 0.005"
wall -	0.045 +/- 0.005"
height	8.685 + 0.000", - 0.005"

To facilitate fitment of the bottom endcap, a pedistal fixture will be used to locate the endcap flush with the tube end. This endcap will be fabricated and welded in place by the ESA/WMA Main Machine Shop. The top endcap will be pressed into place to a flush fit with the tube end. The top endcap will be welded in place by NMT-11.

3) Endcaps for each end of the inner and outer tubes, are required. The endcaps shall be machined from 304 SS flat or bar stock. The dimensions for the inner and outer tube bottom endcaps shall be as specified below and will be fabricated by the ESA/WMA Main Machine Shop. The inner and outer cylinder top endcap configuration and dimensions are as specified in Figure 3. These endcaps will be fabricated by the designated commercial machine shop. An illustration detail of the top inner and outer top endcaps in the assembled configuration is also provided for clarity, see Figure 4.

a) Outer Tube Bottom Endcap Dimensions

O.D. (outer) -	clearance fit to final tube I.D.
thickness -	0.105 +/- 0.007"

b) Outer Tube Top Endcap Dimensions

(Refer to Figure 3)

c) Inner Tube Bottom Endcap Dimensions

O.D. (outer) -	Clearance fit to final tube I.D.
thickness -	0.105 +/- 0.007"

d) Inner Tube Top Endcap Dimensions

(Refer to Figure 3)

Materials Certificates of the 304 seamless stainless steel stock are required for all components; tubing and endcaps.

Machined Part Cleaning/Fabrication

The inner and outer SS seamless tube and associated endcaps are to be machined to specification with burrs and sharp edges removed. The machined components are to be cleaned of any surface residues by the commercial machine shop. After cleaning and drying, the SS components are to be inspected for cleanliness and sealed in plastic bags for delivery to ESA/WMA for receiving inspection, endcap welding, and leak checking.

One endcap (the bottom endcap) is to be TIG or laser welded to one end of each cylinder, inner and outer. The weld is to be helium leak tested to prove leak free at 1×10^{-8} atm-cc/second. Certification of leak testing for each inner and outer tube/bottom endcap assembly is to be provided.

The second (top) endcap of the inner cylinder is to be machined to an O.D. allowing a light interference fit (ANSI Standard Interference Locational Fit, Class LN1) with respect to the inner tubing assembly.

The inner welded tube/endscap assembly is to be fitted into the outer welded tube/endscap assembly prior to packaging to ensure a smooth, unobstructed insertion and seated fit. Since the clearance between the inner and outer cylinders is minimal, it is critical that the cylindricity (roundness) and straightness of the cylinders be controlled to ensure an unobstructed insertion of the inner tube into the outer after the bottom endcaps are welded in place. The inner assembly must set flatly on the bottom of the outer assembly with the top inner cylinder edge being 0.210 ± 0.007 " shorter than the outer tube top edge when fitted together, see Figure 2. When installed, the uppermost edge of the top endcap is to be flush within 0.010" of the outer cylinder top edge and there is to be no perceptible movement of the inner tube assembly when seated, see Figures 2 and 4.

The inner and outer cylinder/endscap assemblies are to be identified as mated components and packaged in a sealed plastic bag for contamination prevention and transportation.

All component external surface finish shall correspond to 63RMS or better.

Assembly Identification.

Each completed cylindrical assembly must have a unique identifier which is a physical attribute of the material, i.e. an indelible identification mark. This shall be accomplished through marking the below listed identification marks on the exterior surface of the inner and outer cylinder top and bottom endcaps. The completed assembly will therefore have a visible serial number imprinted into each outer endcap. The size/font specification shall provide an easy to read label extending across at least 60% of the endcap diameter. The assembly identification marks as of 8/13/98 shall be as follows:

Identification Marks: WRAP1mgPu, WRAP2mgPu, WRAP5mgPu,
WRAP10mgPu, WRAP20mgPu, WRAP50mgPu, WRAP100mgPu,
WRAP200mgPu, WRAP500mgPu, WRAP1.0gPu, WRAP2.0gPu, WRAP5.0gPu,

WRAP10gPu, WRAP20gPu, WRAP30gPu, WRAP40gPu, WRAP50gPu, WRAP1.0gU, WRAP10gU, WRAP20gU, WRAP50gU.

The remaining endcaps shall be delivered without identification marks.

Quantity

The work scope consists of TIG or laser welding one inner endcap to one end of the inner seamless tube and the welding of one outer endcap to one end of the outer seamless tube. Each unit, inner and outer, also requires another machined top endcap to allow the closure of each seamless tube at another facility. The closure of the inner cylinder through welding of the top inner endcap, insertion of this cylinder into the outer followed by welding of the top outer endcap yields the completed assembly. One hundred such assemblies are required which are comprised of the following components:

- 100 inner seamless tubes with welded endcaps
- 100 inner tube top endcaps,
- 100 outer seamless tubes with welded endcaps
- 100 outer tube top endcaps.

An additional 25 each top endcaps shall also be fabricated by the commercial machine shop and require receiving inspection by the ESA/WMA Main Machine Shop.

Packaging

The cleaned and welded assemblies and endcaps shall be packaged in sealed plastic bags such that they are isolated from contaminants. A package is to consist of one complete set of components, i.e. 1 top inner cylinder endcap, 1 top outer cylinder endcap, 1 inner welded tube/encap assembly, and 1 outer welded tube/encap assembly. The inner and outer welded assemblies should be nested together however the endcaps shall be wrapped in an individual plastic bag which in turn shall be placed in the larger plastic bag with the matching welded tube nested assembly. Each single larger bag will contain a set of four mated components and is to be externally identified (black flow pen) by the serial number of the enclosed set. Thus there are to be a total of one hundred large bags each containing two individually wrapped endcaps and a nested inner and outer welded tube/encap assemblies. Additionally, the 25 each inner and outer 'extra' endcaps shall be packaged in plastic bags, no markings required.

Quality Assurance and Control Requirements

ESA/WM shall operate under a Quality Assurance Program that will ensure the one hundred assemblies and 50 extra endcaps meet all technical specifications and are delivered in a cost-effective and timely manner. The Quality Assurance program shall provide controls over procurement, training, certification, safety, operations, inspection, testing, and records.

Specific elements of the ESA/WM Quality Assurance Program are as follows:

- a) QAPjP: ESA/WM shall operate under the CST-8 QAPjP Plan for Non-destructive Assay Performance Demonstration Program Working Reference Material Production, PLA-STD-100, or an alternative plan approved by the NMT-1 Project Manager.
- b) Training and Certification:
 - i) Only Journeyman Prototype Machinists shall perform machining operations. These individuals shall have a minimum of 10 years of industrial machining experience or have successfully completed a four year LANL (or equivalent) Apprentice Machinist program.
 - ii) Only a welder certified for general TIG welding shall perform TIG welding operations on the assemblies. If laser welding is employed, the welder shall be certified through the successful welding and metallographic inspection of stainless steel test items.
 - iii) Helium leak checks shall be performed by a qualified person using a testing instrument calibrated each day of use with a Calibrated Standard leak.
 - iv) Dimensional measurement inspection instruments (tools, gages, instruments) shall only be used for final measurement if they are under active certification by the LANL Standards and Calibration department
- c) Records and Documentation: ESA/WM shall retain and make available as requested, training, educational, qualification and certification records on machinist, welder, and leak check personnel participating in this job. ESA/WM shall submit all records, certificates, and completed forms to NMT-1 for review, filing, storage and eventual delivery to external

customers. These records shall include:

- i) Purchase Order and Materials Certificate (chemical and physical properties, material lot/heat number) for the SS tubing, bar, and sheet stock.
 - ii) Floor Job Order Forms
 - ii) Dimensional measurement inspection forms (as received components and final welded assembly components)
 - iv) Helium Leak Check Weld Inspection forms
 - v) First Assembly Inspection form.
- d) Handling Storage and Shipping: Handling, storage, cleaning, packaging, shipping, and preservation of SS assemblies, endcaps, and test components shall be controlled to prevent damage, contamination, loss, or deterioration.
- e) Control of Nonconforming Items: Items that do not conform to specified requirements shall be controlled to prevent inadvertent installation or use. Controls shall provide for the following:
- * Identification
 - * Documentation
 - * Evaluation
 - * Segregation
 - * Disposition of uncorrectable, nonconforming items
 - * Notification to supervision
- f) Corrective Action: Conditions adverse to quality shall be identified promptly and corrected as soon as practical. In the case of a significant condition adverse to quality, the cause of the condition shall be determined and corrective action taken to preclude recurrence.

Delivery Time

The set of 21 completed assemblies with identification numbers and the 50 additional top endcaps shall be delivered to the NMT-1 Project Manager by ESA/WM within 10 working days after receipt from the tube and endcap components from the commercial machine shop. The final set of 79 completed assemblies shall be delivered to the NMT-1 Project Manager within 60 working days after receipt from the tube and endcap components from the commercial machine shop.

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Tig Or Laser Welded. 100% Penetration To Tube Wall Thickness

Figure 1. Stainless Steel Double Encapsulation

Figure 2. PDP Standard Configuration

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Outer Cylinder Top End Cap
(Style A)

Inner Cylinder Top End Cap
(Style B)

• Approximate wall thickness. Machine to match actual tube wall thickness.

Figure 3. Top Inner and Outer End Cap Dimensions

REVISION to
Fig 3, FLA-STD-103

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Figure 4. Top Inner and Outer End Cap Assembly Detail

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Radiochemical Analysis on Working Reference Materials (WRMs)

for Lawrence Livermore National Laboratory (LLNL)

(June 1998)

A Summary Report

By

**Amy Wong
Denise Thronas**

LANL Analytical Chemistry Group, NMT-1

October 1998

Radiochemical Analysis on Working Reference Materials (WRMs) for LLNL

Analytical Analysis of Starting Materials:

The plutonium oxide (PuO_2) used in preparation of LLNL WRM was PEOR3257B/C/D. The same batch of materials (PEOR3257S) was assayed and used by other projects. The plutonium and americium information is shown below.

Information on Pu Oxide

Plutonium oxide, PEOR 3258B/C/D (LIMS #200021939, 10/96) was used as the starting material for the WRM preparation.

Plutonium Assay: 87.915 ± 0.081 % at 95 % confidence bound on October 2, 1996.
 87.897 ± 0.081 % at 95 % confidence bound on June 25, 1998.

Plutonium Isotopic Composition

Table I.

Nuclide	Half-Life (Years)	Weight percent as of 10/2/96	Weight percent as of 6/25/98
Pu-238	87.74	0.0151	0.0149
Pu-239	24119	92.9661	92.9845
Pu-240	6560	6.7202	6.7206
Pu-241	14.34	0.2327	0.2141
Pu-242	387,000	0.0659	0.0659
Am-241	432.2	0.1114	0.1140

Total Alpha Activity per gram of Plutonium

Table II.

Date	Alpha Activity (mCi/g)
10/2/1996	79.3075
01/01/1997	79.3951
07/01/1997	79.5658
01/01/1998	79.7348
07/01/1998	79.8967
01/01/1999	80.0568
07/01/1999	80.2102
01/01/2000	80.3619
07/01/2000	80.5079
01/01/2001	80.6516
07/01/2001	80.7891
01/01/2002	80.9251
07/01/2002	81.0553

²⁴¹Americium Concentration (μg Am/g Pu)

Table III

Date	μg ²⁴¹ Am /g Pu
10/02/1996	1114
01/01/1997	1141
07/01/1997	1195
01/01/1998	1248
07/01/1998	1299
01/01/1999	1349
07/01/1999	1398
01/01/2000	1446
07/01/2000	1492
01/01/2001	1538
07/01/2001	1582
01/01/2002	1626
07/01/2002	1667

Determination of ²⁴¹Am in the Waste Items by Comparative Method:

Preparation of Waste Item Standard

The same waste item standard which was prepared for the PDP Phase II NDA Standard Preparation was utilized for the LLNL waste item measurements. The simulated waste container was prepared into a standard by taping four cloth swipes spiked with ²⁴¹Am standard solution. These four cloths were arranged symmetrically around the inner wall of the blending bottle. The waste item calibration standard was prepared using the same constituents of an actual waste item; a clean blending bottle, kraft papers, Q-tips, paper funnel, and kimwipes.

The amounts of activity (as of 6/25/98) in the standards were 2.692 μCi for medium activity level standard (labeled as NDA Med. STD, 4/97) and 26.964 μCi for high activity level standard (labeled as NDA High STD, 4/97). The medium activity level standard was prepared from the Amersham ²⁴¹Am solution standard (AMP10040, 95/241/49) (see RC-AW-5, p,43). The high activity level standard was prepared from a pure ²⁴¹Am standardized solution (LIMS#200001684). The Amersham ²⁴¹Am solution is traceable to both NIST and NPL (U.K.). The ²⁴¹Am solution is standardized using calibrated liquid scintillation and gamma counters. The calibrations of these instruments were performed using NIST traceable standards.

Counting Conditions

The amount of ²⁴¹Am in each of the waste items was determined by gamma-ray spectrometry using comparative method. The total number of counts in the ²⁴¹Am 59.6-

keV peak acquired in a fixed time period for a waste item was compared to the total number of 59.6-keV counts for the standard. The waste items and standard bottles were all triply bagged in order to prevent possible contamination.

The conditions for counting (6/22-6/25/98)

- (1) Count time: 600-1800 seconds
- (2) Distance to detector surface: 45 cm
- (3) Background: run 3 times
- (4) Standards: run 4 times each, high and medium standards
- (5) Waste items: run once

Standard Positioning Measurements:

The waste item standard was placed in a fixed counting geometry and was counted four times between 6/22/98 and 6/25/98. The standard bottle was positioned vertically with the center aligned with the axial of the detector.

Uncertainties:

- (1) Positioning and counting statistics:

Medium Standard

Average counts per 1800 seconds: 3320 ± 274 counts, % RSD = 2.1

Counting error at this count rate: 2.0 % (determined by software)

High Standard

Average counts per 1200 seconds: 86587 ± 2591 counts, % RSD = 3.0

Counting error at this count rate: 0.8 % (determined by software)

- (2) Positioning of the waste item:

Positioning term of replacing waste item to the same counting geometry was not determined in this project. The estimate uncertainty for this term was 3 % (obtained from the previous project).

Counting statistics of the waste item measurements:

Each waste item was measured once in a fixed counting geometry with the high purity germanium detector. The counting statistics of the waste item measurements ranged from 0.74 to 1.89 %. The total number of 59.6-keV counts in each waste item was compared to the total number of 59.6-keV counts in the waste item standard and converted to μg of ^{241}Am .

Plutonium content left in the waste item:

The amount of plutonium left in the waste item was calculated from the amount of ^{241}Am found in the blending bottle. As of October 2, 1996, ^{241}Am concentration in plutonium was 1114 ppm (or 979 ppm in PuO_2). After decay and in-growth of ^{241}Am from ^{241}Pu corrections, as June 25, 1998, ^{241}Am concentration in plutonium was 1297 ppm (or 1140 ppm in PuO_2).

The amount of ^{241}Am (μCi) left in each waste bottle was converted to μg of ^{241}Am and then converted to μg of plutonium left in the waste item.

Table IV. LLNL Waste Items INFORMATION

Item	DE, g	as of 6/25/98, in Blend			in Waste Item	
		g, PuO_2	g, Pu	μg , ^{241}Am	μg , ^{241}Am	% left
LLWPU0.1G	133.99	0.11640	0.10231	133	0.585	0.44
LLWPU1.0G	133.52	1.1358	0.99833	1295	0.840	0.06
LLWPU10.0G	131.47	11.37627	9.99940	12969	2.711	0.02
LLWPU25.0G	129.80	28.44401	25.00143	32427	5.889	0.02
LLWPU65.0G	128.08	73.9536	65.00300	84309	11.249	0.01
LLWPU67.0G	127.98	76.2028	66.97998	86873	14.051	0.02
LLWPU68.0G	127.05	77.3546	67.99237	88186	12.733	0.01

LLNL Waste Item Measurements

The net plutonium content in the working reference material:

The net amount of plutonium (as of 6/25/98) in each WRM was calculated as follows:

$$\text{Net Pu, g} = \frac{\text{g of PuO}_2 \text{ in WRM}}{0.87897} - \frac{\mu\text{g of } ^{241}\text{Am in waste item}}{1297 \mu\text{g of } ^{241}\text{Am/g Pu}}$$

Item	Net Amount in WRM	
	g Pu	g PuO2
LLWPU0.1G	0.10186	0.11589
LLWPU1.0G	0.99769	1.13506
LLWPU10.0G	9.99731	11.3739
LLWPU25.0G	24.9969	28.4388
LLWPU65.0G	64.9943	73.9437
LLWPU67.0G	66.9691	76.1905
LLWPU68.0G	67.9826	77.3434

*Decay-corrected Pu assay value = 87.897 % on 6/25/98.

The total alpha activity in WRMs:

The total alpha activity (as of 6/25/98) is calculated based on the alpha specific activity of the plutonium (mCi/g) * grams of plutonium in each WRM. (Plutonium includes plutonium isotopes and ²⁴¹Am isotope.)

$$\text{Total Alpha Activity in each WRM, mCi} = \frac{\text{mCi alpha activity}}{\text{g of Pu}} * \text{g of Pu in WRM}$$

WRM#	Total Alpha Activity, mCi (6/25/98)
LLWPU0.1G	8.138
LLWPU1.0G	79.71
LLWPU10.0G	798.8
LLWPU25.0G	1997
LLWPU65.0G	5193
LLWPU67.0G	5351
LLWPU68.0G	5432

*Pu alpha specific activity = 79.8967 mCi/g on 6/25/98.

Homogeneity Measurement of ^{239}Pu Content within Each Working Reference Material:

Non-destructive gamma-ray analyses were performed on all seven LLWPU working reference materials (WRMs) using a HPGe detector. The purpose of this measurement was to provide a verification of the homogeneity of the Pu content within each WRM. The homogeneity of each WRM was verified by comparing the ^{239}Pu and ^{241}Am count rate of two segments of WRM.

Counting Geometry:

A portable HPGe photon detector was shielded with lead brick with the detector surface facing horizontally. A few lead bricks were used to create a window of 6 cm wide and 65 cm away from the detector surface (Figure I). The WRM was placed horizontally inside of the lead bricks with the left or right side exposed the window opening.

Figure I.

Each side (left or right) was counted for 600 to 10800 seconds. The net areas of gamma-ray peaks at 59.5 keV (^{241}Am), and 129, 375 and 414 keV (^{239}Pu) were recorded.

The Table V showed that all the WRM left to right ratios were close to ratio of 1.0 for all the Pu and Am peaks. This measurement verified that the plutonium content was homogeneous within a WRM.

Verification of ^{239}Pu Content of the Working Reference Materials:

Using the same homogeneous measurement data, we verified the plutonium contents based on the normalized count rate (counts per minutes per gram of Pu). The peak areas of ^{241}Am and ^{239}Pu peaks were analyzed and normalized with the corrected Pu mass. Since we did not perform self-absorption and attenuation corrections on these WRMs, the normalized count rate for different mass groups of WRMs were different (Figures II – III).

Table V. Homogeneous Measurements on LLNL WRMs:

WRM ID	Note	PuO ₂ , g	241Am at 59.5 keV				238Pu at 129 keV			
			Count	Err	cpm	L/R	Count	Err	cpm	L/R
LLWPU0.1G	L	0.114	112734	0.68	626		9549	3.88	53	
LLWPU0.1G	R	0.114	118994	0.67	661	0.95	9832	3.8	55	0.97
LLWPU1.0G	L	1.14	170111	0.54	5670		15478	1.95	516	
LLWPU1.0G	R	1.14	166262	0.54	5542	1.02	15068	1.99	502	1.03
LLWPU10.0G	L	11.37	406864	0.34	40686		39403	1.16	3940	
LLWPU10.0G	R	11.37	418280	0.34	41828	0.97	40206	1.15	4021	0.98
LLWPU25.0G	L	28.44	546251	0.31	54625		72266	0.89	7227	
LLWPU25.0G	R	28.44	542603	0.31	54260	1.01	70279	0.93	7028	1.03
LLWPU65.0G	L	73.94	656564	0.29	65656		94576	0.82	9458	
LLWPU65.0G	R	73.94	684070	0.27	68407	0.96	94528	0.82	9453	1.00
LLWPU65.0G	L	73.94	693870	0.27	69387		94628	0.82	9463	
LLWPU65.0G	R	73.94	688339	0.27	68834	1.01	94540	0.82	9454	1.00
LLWPU67.0G	L	76.21	686729	0.27	68673		94875	0.81	9488	
LLWPU67.0G	R	76.21	680765	0.28	68077	1.01	94839	0.86	9484	1.00
LLWPU68.0G	L	77.35	686155	0.27	68616		94591	0.82	9459	
LLWPU68.0G	R	77.35	686865	0.27	68687	1.00	95076	0.82	9508	0.99
LLWPU68.0G	L	77.35	689214	0.27	68921		95842	0.82	9584	
LLWPU68.0G	R	77.35	680089	0.28	68009	1.01	95355	0.8	9536	1.01

STD ID	Note	PuO ₂ , g	239Pu at 375 keV				239Pu at 413 keV			
			Count	Err	cpm	L/R	Count	Err	cpm	L/R
LLWPU0.1G	L	0.114	2493	7.56	14		2337	7.38	13	
LLWPU0.1G	R	0.114	2433	7.76	14	1.02	2450	7.1	14	0.95
LLWPU1.0G	L	1.14	4119	3.69	137		3802	3.74	127	
LLWPU1.0G	R	1.14	3977	3.72	133	1.04	3604	3.79	120	1.05
LLWPU10.0G	L	11.37	12141	1.98	1214		11381	1.97	1138	
LLWPU10.0G	R	11.37	12619	1.95	1262	0.96	11927	1.93	1193	0.95
LLWPU25.0G	L	28.44	29760	1.28	2976		27904	1.28	2790	
LLWPU25.0G	R	28.44	27758	1.32	2776	1.07	26020	1.32	2602	1.07
LLWPU65.0G	L	73.94	63715	0.9	6372		62636	0.89	6264	
LLWPU65.0G	R	73.94	62987	0.9	6299	1.01	61309	0.9	6131	1.02
LLWPU65.0G	L	73.94	63656	0.9	6366		62042	0.89	6204	
LLWPU65.0G	R	73.94	63519	0.9	6352	1.00	61317	0.9	6132	1.01
LLWPU67.0G	L	76.21	66345	0.87	6635		64079	0.88	6408	
LLWPU67.0G	R	76.21	65249	0.88	6525	1.02	62569	0.89	6257	1.02
LLWPU68.0G	L	77.35	66684	0.87	6668		64223	0.88	6422	
LLWPU68.0G	R	77.35	65325	0.88	6533	1.02	63248	0.89	6325	1.02
LLWPU68.0G	L	77.35	65817	0.88	6582		63434	0.88	6343	
LLWPU68.0G	R	77.35	66234	0.87	6623	0.99	64011	0.88	6401	0.99

Figure II. Normalized Count Rate for ^{241}Am 59.5 keV peak:

Figure III. Normalized Count Rate for ^{239}Pu 414-keV peak:

Final Certificate Values with Uncertainties (as of 6/25/98):

Table VI gives the final Pu, PuO₂, ²⁴¹Am, and total alpha activity (mCi and Bq) values with estimated uncertainty in each WRM.

WRM#	Mass in WRM, as of 6/25/98					
	g, Pu	± Pu, g	g, PuO ₂	± PuO ₂ , g	mg, ²⁴¹ Am	± Am, mg
LLWPU0.1G	0.10186	0.00014	0.11589	0.00016	0.1321	0.0009
LLWPU1.0G	0.99769	0.00098	1.13506	0.00112	1.2940	0.0086
LLWPU10.0G	9.99731	0.00938	11.3739	0.0107	12.967	0.086
LLWPU25.0G	24.9969	0.0234	28.4388	0.0266	32.421	0.215
LLWPU65.0G	64.9943	0.0606	73.9437	0.0689	84.298	0.559
LLWPU67.0G	66.9691	0.0626	76.1905	0.0712	86.859	0.576
LLWPU68.0G	67.9826	0.0634	77.3434	0.0722	88.173	0.584

WRM#	Total Alpha, as of 6/25/98			
	mCi	±, mCi	Bq	±, Bq
LLWPU0.1G	8.138	0.023	3.011E+08	9E+05
LLWPU1.0G	79.71	0.23	2.949E+09	9E+06
LLWPU10.0G	798.8	2.3	2.955E+10	8E+07
LLWPU25.0G	1997	6	7.390E+10	2E+08
LLWPU65.0G	5193	15	1.921E+11	6E+08
LLWPU67.0G	5351	15	1.980E+11	6E+08
LLWPU68.0G	5432	16	2.010E+11	6E+08

Uncertainty Calculation:

- Pu Isotopic Analysis***

Isotopic Analysis (10/2/96)

Isotope	wt % (10/96)	SD	Measurement*		Table of Radioactive Isotopes			
			%RSD	At 95 % ± wt %	SA, Uncertainty %	%α	Uncertainty In Alpha activity, %	Unc. In Pu sample, %
²³⁸ Pu	0.01508	0.00037	2.4545	± 0.000005	0.1026	100	2.457	0.00037
²³⁹ Pu	92.9661	0.001227	0.0013	± 0.000015	0.1244	100	0.124	0.11566
²⁴⁰ Pu	6.7202	0.000561	0.0084	± 0.000007	0.1067	100	0.107	0.00719
²⁴¹ Pu	0.2327	0.000778	0.3343	± 0.000010	1.389	0.00241	0.000	0.00000
²⁴² Pu	0.06592	0.000249	0.3777	± 0.000003	0.531	100	0.652	0.00043
²⁴¹ Am	0.09792	0.000217	0.2214	± 0.000002	0.116	100	0.250	0.00024
SUM								0.1239

*Measurement % RSD is based on the duplicated analysis of 5 cuts of sample. The uncertainty at 95% CL is based on the long and short terms uncertainty.

- Pu Assay***

Pu assay value was 0.87915 ± 0.00064 . At 95 % CL, the standard deviation = 0.00081.

- Am Assay***

Am assay value based on mass spectrometry data was 979.2 ± 0.2 µg/g. At 95 % CL, % of RSD was 0.27 %. With the uncertainty (0.25 %) of isotope dilution mass spectrometric method, the total uncertainty for Am Assay is ± 0.52 % at 95 % CL.

Radiochemistry also determined ²⁴¹Am concentration in the PuO₂ sample by liquid scintillation analysis and gamma-ray spectrometry. The liquid scintillation counter (Packard Tri-Carb 2200C) was calibrated using an NIST traceable Radiochemistry Primary plutonium oxide standard (PEOR3258-STD1) The NaI based gamma-ray spectrometer was calibrated using an NIST traceable ²⁴¹Am standard solution (Amersham, AMP10040, batch 95/241/49). The ²⁴¹Am concentration was 980.8 µg/g of PuO₂ (% RSD = 0.55 %).

- **Weighing Error:**

Wt range	AVG	1 SD	at 95% CL	
			SD	% RSD
20 mg	0.019998	5.00E-06	0.000016	0.0800
100 mg	0.100015	0.000008	0.000022	0.0220
5 g	5.000037	0.000011	0.000027	0.0005
10 g	10.00005	0.000079	0.000019	0.0002
20 g	20.00002	0.000021	0.000045	0.0002
50 g	50.00016	0.000075	0.00016	0.0003
100 g	100.0002	0.000139	0.00032	0.0003

- **Alpha Activity:**

$$\text{Activity (mCi)} = \text{Specific Activity (mCi/g)} * \text{Weight (g)}$$

Based on the uncertainties of Pu isotopic composition and Pu assay, the uncertainty of Pu specific activity is 0.287 % at 95 % CL. For each weight range, the uncertainty = $\sqrt{(\text{uncertainty of specific activity})^2 + (\text{uncertainty of weight range})^2}$ at 95 % CL.

- **Waste Retention Uncertainty Calculation:**

During the LLNL NDA WRM Preparation, the waste item hold-up measurements were performed by gamma-ray analysis using a portable HPGe detector. The measured count rate (cpm) of the waste item was compared to the average normalized count rate (cpm/ μ Ci) obtained from the standards. Two standards were prepared from ^{241}Am standardized solution and Amersham ^{241}Am solution standard.

The error terms associated with each process of preparing the waste item standards and measuring the waste items are listed in the next page. Each error term was identified either as random or systematic error.

The total errors associated with waste item measurements (relative bias of Am measurements) were used in the Pu and Am content calculation. The total error was estimated individually based on the count rate of the measured ^{241}Am peak area for each waste item. Low count rate (cpm) has higher percent of error associated with it.

Table VII gives comprehensive uncertainties calculation of waste items.

LLNL: Error Estimate for the Waste Item Hold-Up Measurements

In the NDA LLNL WRM Production, the waste item hold-up measurements were performed by gamma-ray analysis using a portable HPGe detector. The measured count rate (cpm) of the waste item was compared to the average (cpm/ μCi) obtained from the standards. Two standards were prepared from ^{241}Am standardized solution and Amersham ^{241}Am solution standard.

The following error terms were identified and estimates of random and systematic errors listed:

Error Term	n	Random	Systematic	Note
<i>Med. Level Standard Preparation</i>				
Concentration of Amersham ^{241}Am solution standard, total uncertainty	1			certificate
Gravimetric dilution for a 50-g solution	1	0.00015%	0.50%	BM wt data
Pipetting Amersham ^{241}Am solution standard on the filters	4	max 0.5 %	max 0.5 %	
<i>High Level Standard Preparation</i>				
Concentration of Amersham ^{241}Am solution standard, total uncertainty	1			certificate
Weighing ~0.5 g of ^{241}Am Amersham solution as a calibration standard	5	0.0080%	0.50%	BM wt data
Counting efficiency determination of the gamma counter	10	0.23%		%RSD
Pipetting ^{241}Am solution for standardization	3	max 0.5 %	max 0.5 %	
Standardization of ^{241}Am on the gamma counter	3	0.11%		%RSD
Pipetting standardized ^{241}Am solution on the filters	4	max 0.5 %	max 0.5 %	
<i>Position/Rotation of the waste item standard</i>	4		max 1.2 %	positioning
<i>Attenuation, variation from standard to waste item</i>	1			Based on 1 vs 1.5 mm bottle wall thickness. T=99 vs 98%
<i>Average cpm per μCi</i>	5	0.58%	max 1 %	%RSD
<i>Calibration Curve, linearity (the slope, cpm per μCi)</i>	1		0.10%	regression curve

LLNL: Error Estimate for the Waste Item Hold-Up Measurements

In the NDA LLNL WRM Production, the waste item hold-up measurements were performed by gamma-ray analysis using a portable HPGe detector. The measured count rate (cpm) of the waste item was compared to the average (cpm/ μ Ci) obtained from the standards. Two standards were prepared from ^{241}Am standardized solution and Amersham ^{241}Am solution standard.

The following error terms were identified and estimates of random and systematic errors listed:

<i>Calibration Curve, zero offset (< 1 cpm)</i>		
<i>Estimated error obtained from the calibration curve</i>		
327 cpm		
469 cpm		
1514 cpm		
3289 cpm		
6281 cpm		
7846 cpm		
7110 cpm		
	negligible	
		Estimated from regression curve
	1.36%	0.798%
	0.94%	0.531%
	0.29%	0.143%
	0.13%	0.094%
	0.07%	0.099%
	0.05%	0.102%
	0.06%	0.100%

The combined error terms were combined based on random and systematic errors.

Random Errors, % = $\text{Sqrt}((0.00015)^2 + (0.5)^2 + (0.008)^2 + (0.23/\text{sqrt}(10))^2 + (0.5)^2 + (0.11/\text{sqrt}(3))^2 + (0.5)^2 + (0.58/\text{sqrt}(5))^2 + (\text{calib. curve})^2)$

327 cpm	1.640 %
469 cpm	1.307
1514 cpm	0.953
3289 cpm	0.919
6281 cpm	0.912
7846 cpm	0.911
7110 cpm	0.911

LLNL: Error Estimate for the Waste Item Hold-Up Measurements

In the NDA LLNL WRM Production, the waste item hold-up measurements were performed by gamma-ray analysis using a portable HPGe detector. The measured count rate (cpm) of the waste item was compared to the average (cpm/ μ Ci) obtained from the standards. Two standards were prepared from ^{241}Am standardized solution and Amersham ^{241}Am solution standard.

The following error terms were identified and estimates of random and systematic errors listed:

Systematic correction, multiplying $(1 + \text{bias term}) = (1 + 0.005) * (1 + 0.005) * (1 + 0.005) * (1 + 0.012) * (1 + 0.01) * (1 + \text{calib. curve})$				
			Total error = systematic bias + 2 * (random bias)	
327 cpm	1.057	5.73%	7.37 Total, %	9.01
469 cpm	1.055	5.45%	6.76	8.07
1514 cpm	1.050	5.05%	6.00	6.95
3289 cpm	1.050	5.00%	5.92	6.83
6281 cpm	1.050	5.00%	5.91	6.82
7846 cpm	1.050	5.00%	5.92	6.83
7110 cpm	1.050	5.00%	5.91	6.83
1 Sys + 1 Random				

H

PDP Standard Information
(Statistical Calculation)

11/12/98

	A	B	C	D	E	F	G	H	I	J	K	L	M
1			as of 10/96	as of 6/25/98	as of 10/96	as of 6/25/98	as of 6/25/98	%Ct err			net	net	net
2			g Pu	g Pu	µg Am	µg Am	µg Am	for Am		% loss	Am241	Pu	Pu
3	WRM#	gPuO2	87.915%	87.897%	1113.72	1297.0	loss	RSD	cpm		6/98, µg	6/98, g	10/96, g
4	LLWPU0.1G	0.11640	0.10233	0.10231	114	133	0.585	0.940	327	0.441	132	0.10186	0.10188
5	LLWPU1.0G	1.13580	0.99854	0.99833	1112	1295	0.840	1.890	469	0.065	1294	0.99769	0.99789
6	LLWPU10.0G	11.37627	10.00145	9.99940	11139	12972	2.711	1.240	1514	0.021	12969	9.99731	9.99936
7	LLWPU25.0G	28.44401	25.00666	25.0014	27850	32433	5.889	1.180	3289	0.018	32428	24.9969	25.0020
8	LLWPU65.0G	73.9536	65.0163	65.0030	72410	84326	11.249	0.850	6281	0.013	84315	64.9943	65.0076
9	LLWPU67.0G	76.2028	66.9937	66.9800	74612	86891	14.051	0.760	7846	0.016	86877	66.9691	66.9829
10	LLWPU68.0G	77.3546	68.0063	67.9924	75740	88204	12.733	0.800	7110	0.014	88191	67.9826	67.9965
11													
12													
13													
14													
15		Variance of the fraction of Am in PuO2	6.49E-12						1 SD fraction of Am in PuO2				
16		Mean of the fraction of Am in PuO2	9.799E-04							2.55E-06			
17		Variance of the fraction of Pu in PuO2	1.64E-07						1 SD fraction of Pu in PuO2			9.77E-04	
18		Mean of the fraction of Pu in PuO2	0.87915							0.000405			
19													
20		PuO2 meas. variance	20 mg		2.50E-11								
21			100 mg		6.40E-11								
22			5 g		1.21E-10								
23			10 g		6.24E-09								
24			20 g		4.41E-10								
25			50 g		5.63E-09								
26			100 g		1.93E-08								
27		Rel. Bias Am Meas	327cpm		0.090								
28			469 cpm		0.081								
29			1514 cpm		0.070								
30			3289 cpm		0.068								
31			6281 cpm		0.068								
32			7846 cpm		0.068								
33			7110 cpm		0.068								
34													
35													

Rel. Bias of Am Measurement is estimated from waste measurement and waste standard.

PDP Standard Information
(Statistical Calculation)

11/12/98

	A	N	O	P	Q	R	S	T	U	V	W	X
1		as of 10/96	as of 6/98	PuO2 Bias	PuO2 Loss	Net PuO2	Var[PuO2]	Var[Pu]	Var[Am]	2*%"RSD"	2*%"RSD"	PuO2 Bias
2		α -act, mCi	α -act, mCi	due to	as of 10/96	as of 10/96	(net)			PU	AM	Due to Am Bias
3	WRM#	79.3075	79.8967	Am bias, g	g	g	g	g	g			1.0025 g
4	LLWPU0.1G	8.0800	8.1383	5.385E-05	0.000513383	0.11589	2.5E-11	2.22192E-09	8.72E-14	0.1390	0.4869	5.37E-05
5	LLWPU1.0G	79.1403	79.7119	6.914E-05	0.000736306	1.13506	6.4E-11	2.11354E-07	8.363E-12	0.0983	0.4521	6.90E-05
6	LLWPU10.0G	793.0241	798.7521	1.923E-04	0.00237713	11.37389	1.21E-10	2.12172E-05	8.397E-10	0.0938	0.4483	1.92E-04
7	LLWPU25.0G	1982.8470	1997.1691	4.107E-04	*0.003164976	28.43885	5.625E-09	0.00013265	5.25E-09	0.0936	0.4481	4.10E-04
8	LLWPU65.0G	5155.5930	5192.8321	7.834E-04	0.00986505	73.94373	1.9321E-08	0.000896764	3.549E-08	0.0932	0.4478	7.81E-04
9	LLWPU67.0G	5312.2430	5350.6136	9.788E-04	0.012323066	76.19048	1.9321E-08	0.000952086	3.768E-08	0.0934	0.4480	9.76E-04
10	LLWPU68.0G	5392.6308	5431.5820	8.869E-04	0.011166464	77.34343	1.9321E-08	0.000981119	3.883E-08	0.0933	0.4479	8.85E-04
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PDP Standard Information
(Statistical Calculation)

11/12/98

A	Y	Z	AA	AB	AC	AD	AE	AF	AG	AH
	PuO2 Bias ue to Am Bia	Var[PuO2] (net)	Var[PuO2] (net)	Var[Am] g	Var[Am] g	gAm	gAm	plus 2sd Am	minus 2sd Am	Diff / 2 / gAm 95% bound
1										
2										
3	WRM#	0.9975 g	0.9975 g	g	g	1.0025	9.975000e-01			
4	LLWPU0.1G	5.40E-05	5.63758E-11	8.71961E-14	8.71958E-14	1.138960e-04	1.132230e-04	0.000114487	0.000112632	0.7016
5	LLWPU1.0G	6.93E-05	3.24914E-10	8.36284E-12	8.36284E-12	1.115118e-03	1.109421e-03	0.001120901	0.001103638	0.6669
6	LLWPU10.0G	1.93E-04	1.29159E-09	8.39708E-10	8.39708E-10	1.117353e-02	1.111743e-02	0.01123149	0.011059476	0.6632
7	LLWPU25.0G	4.12E-04	1.06295E-08	5.2497E-09	5.2497E-09	2.793781e-02	2.779767e-02	0.028082718	0.027652757	0.6630
8	LLWPU65.0G	7.85E-04	2.87941E-08	3.54906E-08	3.54906E-08	7.264071e-02	7.227689e-02	0.07301749	0.071900107	0.6626
9	LLWPU67.0G	9.81E-04	3.11384E-08	3.76801E-08	3.76801E-08	7.484803e-02	7.447281e-02	0.075236257	0.074084587	0.6628
10	LLWPU68.0G	8.89E-04	3.00725E-08	3.88291E-08	3.88291E-08	7.598057e-02	7.559988e-02	0.076374669	0.07520578	0.6627
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PDP Standard Information
(Statistical Calculation)

11/12/98

	A	AI	AJ	AK	AL	AM	AN	AO	AP	AQ	AR
1				Uncertainty due to just biases	Var[Pu] ing max bias effe g	PU uncertainty Using largest bias	at-least 95% Conf. Bound for Pu, %	at-least 95% Conf. Bound for Am, %			
2											
3	WRM#										
4	LLWPU0.1G	9.27E-07	3.37E-07	0.254679467	2.24641E-09	0.139654208	0.140	0.702			
5	LLWPU1.0G	8.63E-06	2.85E-06	0.220067603	2.11538E-07	0.098311931	0.098	0.667			
6	LLWPU10.0G	8.60E-05	2.81E-05	0.216295502	2.12181E-05	0.093846719	0.094	0.663	2.275979		
7	LLWPU25.0G	2.15E-04	7.01E-05	0.216085903	0.000132654	0.093599891	0.094	0.663			
8	LLWPU65.0G	5.59E-04	1.82E-04	0.215754081	0.000896771	0.093212379	0.093	0.663			
9	LLWPU67.0G	5.76E-04	1.88E-04	0.215947131	0.000952096	0.093438212	0.093	0.663			
10	LLWPU68.0G	5.84E-04	1.90E-04	0.215828796	0.000981127	0.093299722	0.093	0.663			
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National Bureau of Standards

Certificate of Analysis

Standard Reference Material 948

Plutonium Isotopic Standard

This Standard Reference Material (SRM) is certified as an isotopic standard for use in isotopic measurements of plutonium. SRM 948 consists of approximately 250 mg of plutonium in the form of plutonium sulfate tetrahydrate packaged in a glass microbottle.

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Atom Percent*	0.010 ± 0.001	91.736 ± 0.010	7.922 ± 0.010	0.299 ± 0.001	0.0330 ± 0.0003

*As of January 1, 1982, refer to Table 1 for quarterly decay-adjusted values.

The plutonium isotopic distribution was determined by thermal ionization mass spectrometry at the National Bureau of Standards (NBS) on samples from which americium and uranium were removed. Because high-purity plutonium isotopes have not been used to prepare known synthetic mixtures, the accuracy is dependent on uranium and plutonium exhibiting similar behavior. The observed mass spectrometer data were corrected for mass discrimination effects using data from the analysis of uranium isotopic SRM's that had been analyzed under similar conditions. In addition, the value for ^{238}Pu was checked by alpha-count of ^{238}Pu , using the known value for ^{239}Pu as a ratio check.

SRM 948 contains uranium and americium isotopes, including growing-in daughters of plutonium that are isobaric with the plutonium isotopes. In addition, there may be radiation damage to the glass bottle and teflon cap, resulting in small glass slivers. Therefore, in its use, a chemical separation that provides a purified plutonium fraction is essential to the attainment of high accuracy.

Measurements leading to the certification of this SRM were made in the Inorganic Analytical Research Division by E.L. Garner, L.A. Machlan, and W.R. Shields.

The technical and support aspects involved in the revision of this Certificate were coordinated through the Office of Standard Reference Materials by T.E. Gills.

August 19, 1982
Washington, D.C. 20234
(Revision of Certificate
dated 12-3-71)

George A. Uriano, Chief
Office of Standard Reference Materials

(over)

U. S. Department of Energy
New Brunswick Laboratory

New Brunswick Laboratory Certified Reference Materials Certificate of Analysis

CRM No. 128

Plutonium-239/Plutonium-242, 1:1 Atom Ratio
In Nitrate Form
(Plutonium Isotopic Standard)

Plutonium-239/Plutonium-242 $0.9993_7 \pm 0.0002_6$ Atom Ratio*

*As of October 1, 1984. Refer to Table I for Yearly Decay-Adjusted Values

This Certified Reference Material (CRM) is primarily intended for the calibration of mass spectrometers used to perform plutonium isotopic measurements. The specific purpose of this isotope standard is for the determination of a mass discrimination factor which will place measured plutonium isotopic ratios on an absolute basis. Each unit of CRM No. 128 consists of approximately 1 mg of a nominal 1:1 mixture of ^{239}Pu and ^{242}Pu , as evaporated plutonium nitrate contained in a 30-mL Teflon bottle. *NOTE: The bottle and its outer plastic containment should be handled under proper radiologically controlled conditions at all times.*

The statistical uncertainty assigned to the certified ratio value is the 95% confidence interval for the unweighted mean of the ratio calculated from assay and mass measurements of the separated isotopes and the ratio determined by mass spectrometric measurements of the CRM. The uncertainty is propagated from all known nonnegligible sources of random and systematic variations associated with the measurement methods used.

The plutonium materials used to produce this CRM were obtained from the ORNL Isotope Sales Group with the approval of the DOE Research Materials/Transplutonium Program Committee chaired by J. L. Burnett. Preparation and assay measurements of the CRM and calibration mixtures were performed by C. G. Cacic, NBL; isotopic measurements were performed by D. W. Crawford, NBL; impurity measurements were performed by J. A. Carter and associates, ORNL. Technical assistance was provided by L. A. Machlan, NBS. Isotopic verification measurements were performed by E. L. Callis, ANL. Statistical assessment of the data for certification was performed by M. D. Soriano, NBL. Initial project technical direction was provided by E. L. Garner, NBS; overall direction and coordination of the preparation, certification and issuance of this CRM were provided by N. M. Trahey, NBL.

October 1, 1985
Argonne, Illinois
(Revision of Certificate,
dated, 10/1/84)

Carleton D. Bingham
Director

(Over)

The ^{239}Pu and ^{242}Pu separated isotopes ($\geq 99.9\%$ isotopic purity) comprising CRM No. 128 were separately dissolved, chemically purified, and assayed by controlled potential coulometry before being combined by weight. The assay characterization measurements were used to calculate a precise gravimetric $^{239}\text{Pu}/^{242}\text{Pu}$ value for the CRM. The CRM was then isotopically equilibrated, apportioned, and dried into units. Isotopic certification measurements were performed on CRM units randomly selected according to a statistical sampling plan. The $^{239}\text{Pu}/^{242}\text{Pu}$ ratio data obtained for the CRM were corrected for mass discrimination effects by concurrent analysis of five $^{239}\text{Pu}/^{242}\text{Pu}$ calibration mixtures, prepared by weight to closely bracket the isotopic ratio of the CRM. Total element impurity content was determined by spark source mass spectrometry on selected subsamples and is estimated to be $300\text{ }\mu\text{g/g}$ plutonium. Although the CRM was americium-free at the time of preparation, the calculated americium ingrowth from the decay of ^{241}Pu present in small amounts in the CRM is $21\text{ }\mu\text{g/g}$ plutonium as of October 1, 1984, and will increase at a rate of approximately 5 percent of the total ^{241}Pu per year.

CRM No. 128 had a radioactivity of $2.7 \times 10^6\text{ Bq}$ ($73\text{ }\mu\text{Ci}$) per unit as of July 1, 1984, which is dominated by ^{239}Pu and ^{241}Pu .

The change with time in the CRM No. 128 certified ratio is very small, due to the relatively long half-lives of the primary plutonium isotopes. Table I provides decay-adjusted values for the certified ratio at one-year intervals for a five-year period.

TABLE I
CRM No. 128 Yearly Decay-Adjusted $^{239}\text{Pu}/^{242}\text{Pu}$ Atom Ratio Value

October 1, 1985	October 1, 1986	October 1, 1987	October 1, 1988	October 1, 1989
0.9993 ₅	0.9993 ₂	0.9992 ₉	0.9992 ₆	0.9992 ₄

The plutonium isotopic distribution of CRM No. 128 is given in Table II and is for informational purposes only (not certified).

TABLE II
CRM No. 128 Isotopic Distribution (as of October 1, 1984)

Plutonium Isotope:	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{244}Pu
Atom Percent:	0.004	49.947	0.035	0.036	49.978	<0.001

(The half-life values used to calculate the values in Tables I and II are expressed in years: ^{238}Pu — 87.74; ^{239}Pu — 24,119; ^{240}Pu — 6,562; ^{241}Pu — 14.35; ^{242}Pu — 376,300.)

RECOMMENDED PROCEDURE FOR USING CRM NO. 128

Each CRM unit contains $1 \pm 0.03\text{ mg}$ of plutonium and is designated for in-situ dissolution. When converted to solution form, a unit can be used as is. No additional purification of the CRM is required.

Wipe the Teflon bottle with a chamois or damp cloth to dissipate any static charge which may cause expulsion of the material upon opening. Unscrew the cap, add 1N HNO_3 to the CRM concentration desired and carefully warm the bottle to insure total dissolution. *Do not heat the bottle above 150°C because bottle deformation will occur!* Replace and tighten the cap, then allow the bottle to cool before shaking to homogenize contents. Wipe cap and bottle threads each time a portion of the CRM solution is removed from the bottle.

Department of Commerce
Malcolm Baldrige
Secretary
National Bureau of Standards
Ernest Ambler, Director

*Manlym - Place f.k.
NDA PDP File # STD. 14
28u*

Mendoza

National Bureau of Standards

Certificate of Analysis

Standard Reference Material **949f**

*Number recently
changed to 126.
28u*

Plutonium Metal

(In cooperation with the University of California Los Alamos
National Laboratory, Los Alamos, New Mexico)

This Standard Reference Material is certified for plutonium content and is primarily intended for use in the chemical assay of plutonium. SRM 949f consists of approximately 0.5 gram of plutonium metal that has been sealed in a glass tube under a reduced-pressure argon atmosphere. Each unit is identified by tube number and sample mass.

Plutonium assay, percent _____ 99.99 (As of October, 1980,
[99.90-100] See Table 2.)

Tube No. _____

Mass _____

^aThe uncertainty of the certified value is expressed at the 99% one-sided confidence interval

The plutonium assays were performed by the Los Alamos National Laboratory with collaborative analysis performed at the Department of Energy, New Brunswick Laboratory. The Los Alamos National Laboratory used two titrimetry methods utilizing both photometric and potentiometric end points [1,2,3,4]. The New Brunswick Laboratory used a controlled-potential coulometric method [5,6]. The certified value is based on the results from these two laboratories using three different methods. However, the confidence interval on the certified value is a one-sided 99% confidence interval on the mean of the potassium dichromate titrimetric method, which is 99.98-100, expanded by the bound on the systematic error ± 0.08 . The 99% confidence interval is based upon the potassium dichromate method because adequate systematic error bounds are available. The mean value for the potassium dichromate titrimetric method is 99.999 with a standard error on the mean of ± 0.006 based on 16 determinations.

The americium resulting from the decay of 14.35 years plutonium-241 was approximately 21 $\mu\text{g/g}$ as of October 1980. The total of all detected impurities is approximately $115 \pm 65 \mu\text{g/g}$. *99.9885*

The technical and support aspects leading to certification and issuance of this Standard Reference Material was coordinated through the Office of Standard Reference Materials by T. E. Gills.

The statistical assessment of the data for the certification of this SRM was performed by W. Liggett of the Statistical Engineering Division of the National Bureau of Standards.

Washington, D.C. 20234
May 11, 1982

George A. Uriano, Chief
Office of Standard Reference Materials

(over)

Supplemental Information

Isotopic Analysis

Plutonium isotopic distribution was determined by thermal ionization mass spectrometry at the Los Alamos National Laboratory. The values given in Table 1 are *not certified*, but given for information only.

Table 1

<u>Isotope</u>	<u>Isotope Mass</u>	<u>Average Atom % on October 8, 1980</u>
²³⁸ Pu	238.04942	0.0040
²³⁹ Pu	239.05203	97.121
²⁴⁰ Pu	240.05382	2.804
²⁴¹ Pu	241.05669	0.065
²⁴² Pu	242.05864	0.006

Calculated Atomic Wt. 239.082

Notice and Warnings to Users

SRM 949f may contain small pieces of Pu metal that may have become separated from the larger pieces in the tube. It is recommended that the tube be carefully rinsed with 3 M hydrochloric acid during the transfer.

The mass of each sample was determined with an overall accuracy of ± 0.05 mg. The assigned mass for each unit, however, has not been corrected for the buoyancy of the dry argon atmosphere in which the plutonium metal was weighed and packaged. The computed atmosphere buoyancy correction factor for dry argon at 592 mm Hg and 20°C is 0.99991 for plutonium metal (having a density of 19.7 g/cm³) relative to brass weights. The value given on this certificate for the mass of the plutonium may be multiplied by this factor to obtain the "true" in vacuo mass.

SRM 949f had a radioactivity of approximately 0.07 Ci per unit, as of October, 1980. The material is believed to be stable and free of oxidation; however, the metal should be inspected before use for signs of oxidation. If oxidation has occurred, the material should be acid-etched, dried, and reweighed before use.

The decay-adjusted values for plutonium in 949f for a 5-year period are shown in Table 2. The half-life values, in years, used for the decay adjustments are ²³⁸Pu, 87.74; ²³⁹Pu, 24119; ²⁴⁰Pu, 6560; ²⁴¹Pu, 14.35; and ²⁴²Pu, 387,000.

Table 2

Plutonium Assay, Percent

<u>1980</u>	<u>1981</u>	<u>1982</u>	<u>1983</u>	<u>1984</u>
99.99	99.98	99.98	99.97	99.97

References

1. C. E. Caldwell, L. S. Griffl, F. J. Miner, and N. E. Moody, *Anal. Chem.* **34**, 346, 1962.
2. G. R. Waterbury and C. S. Metz, Los Alamos Scientific Lab Report, LA 3141, 1965.
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5. M. K. Holland, J. R. Weiss, and C. E. Pietri, *Anal. Chem.* **50**, 236-240 (1978).
6. D. D. Jackson, R. M. Hollen, F. R. Roensch, and J. E. Rein, *Anal. Chim. Acta* **117** 205-15 (1980).

National Institute of Standards & Technology

Certificate

Standard Reference Material 4322B Alpha-Particle Solution Standard

Radionuclide	Americium-241
Source identification	SRM 4322B
Source description	Liquid in NIST borosilicate-glass ampoule ⁽¹⁾
Source mass	Approximately 5.2 grams
Solution composition	Americium-241 in 1 molar nitric acid ⁽²⁾
Reference time	1200 EST September 9, 1991
Radioactivity concentration	39.24 Bq g ⁻¹
Overall uncertainty	0.95 percent ⁽³⁾
Alpha-particle-emitting impurities	None observed ⁽⁴⁾
Photon-emitting impurities (Radioactivity concentration at reference time)	²³³ Pa/ ²⁴¹ Am: $(8.1 \pm 1.6) \times 10^{-4}$ ⁽⁵⁾
Half life	432.2 $\pm$ 0.5 years ⁽⁶⁾
Measuring instrument	4 $\pi\alpha$ liquid-scintillation counter

This standard reference material was prepared in the Physics Laboratory, Ionizing Radiation Division, Radioactivity Group, Dale D. Hoppes, Group Leader.

Gaithersburg, MD
October, 1991

William P. Reed, Chief
Standard Reference Materials Program

*Notes on back

National Bureau of Standards

Certificate of Analysis

Standard Reference Material 946

Plutonium Isotopic Standard

This Standard Reference Material (SRM) is certified as an isotopic standard for use in isotopic measurements of plutonium. SRM 946 consists of approximately 250 mg of plutonium in the form of plutonium sulfate tetrahydrate packaged in a glass microbottle.

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Atom Percent*	0.232 ± 0.007	84.464 ± 0.015	12.253 ± 0.015	2.477 ± 0.005	0.574 ± 0.003

*As of January 1, 1982, refer to Table 1 for quarterly decay-adjusted values.

The plutonium isotopic distribution was determined by thermal ionization mass spectrometry at the National Bureau of Standards (NBS) on samples from which the americium and uranium were removed. Because high-purity plutonium isotopes have not been used to prepare known synthetic mixtures, the accuracy is dependent on uranium and plutonium exhibiting similar behavior. The observed mass spectrometer data were corrected for mass discrimination effects using data from the analysis of uranium isotopic SRM's that had been analyzed under similar conditions.

SRM 946 contains uranium and americium isotopes, including growing-in daughters of plutonium that are isobaric with the plutonium isotopes. In addition, there may be radiation damage to the glass bottle and the teflon cap liner resulting in small glass slivers. Therefore, in its use, a chemical separation that provides a purified plutonium fraction is essential to the attainment of high accuracy.

Measurements leading to the certification of this SRM were made in the Inorganic Analytical Research Division by E.L. Garner and L.A. Machlan.

The technical and support aspects involved in the revision of this Certificate were coordinated through the Office of Standard Reference Materials by T.E. Gills.

August 19, 1982
Washington, D.C. 20234
(Revision of Certificate
dated 12-3-71)

George A. Uriano, Chief
Office of Standard Reference Materials

(over)

The decay-adjusted values for the plutonium isotopic composition, in atom percent, are tabulated below in Table I. The half-life values, in years, used for the decay-adjustment are: ^{238}Pu , 87.74; ^{239}Pu , 24,119; ^{240}Pu , 6,560; ^{241}Pu , 14.34; and ^{242}Pu , 387,000.

Table I
Decay-Adjusted Plutonium Isotopic Composition
Atom Percent

Date	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
January 1, 1982	0.232	84.464	12.253	2.477	0.574
April 1, 1982	.231	84.490	12.257	2.448	.574
July 1, 1982	.231	84.515	12.260	2.419	.575
October 1, 1982	.230	84.540	12.264	2.391	.575
January 1, 1983	.230	84.565	12.267	2.363	.575
April 1, 1983	.230	84.590	12.270	2.335	.575
July 1, 1983	.229	84.614	12.274	2.308	.575
October 1, 1983	.229	84.638	12.277	2.281	.575
January 1, 1984	.229	84.662	12.280	2.253	.576
April 1, 1984	.228	84.685	12.283	2.227	.576
July 1, 1984	.228	84.709	12.286	2.201	.576
October 1, 1984	.227	84.732	12.290	2.175	.576
January 1, 1985	.227	84.755	12.293	2.149	.576
April 1, 1985	.227	84.777	12.296	2.125	.576
July 1, 1985	.226	84.799	12.299	2.100	.577
October 1, 1985	.226	84.821	12.302	2.075	.577
January 1, 1986	.225	84.843	12.305	2.050	.577
April 1, 1986	.225	84.864	12.307	2.026	.577
July 1, 1986	.225	84.885	12.310	2.003	.577
October 1, 1986	.224	84.906	12.313	1.979	.577
95% Confidence Limit:	± 0.007	± 0.015	± 0.015	± 0.005	± 0.003

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NBS-948 Analyses

Instrument: VG-2 Total Evaporation Method Decay Corrected to 10/1/96

Date	Weight %				
	238	239	240	241	242
7/29/96	0.0107	91.8461	7.9596	0.1483	0.0333
8/7/96	0.0163	91.8405	7.9582	0.1497	0.0336
8/13/96	0.0106	91.8466	7.9605	0.1473	0.0335
8/14/96	0.0109	91.8445	7.9616	0.1476	0.0339
8/22/96	0.0103	91.8466	7.9596	0.1486	0.0337
8/26/96	0.0102	91.8457	7.9598	0.1491	0.0341
8/27/96	0.0104	91.8449	7.9600	0.1496	0.0340
8/29/96	0.0107	91.8434	7.9608	0.1503	0.0338
8/30/96	0.0107	91.8434	7.9611	0.1501	0.0338
9/6/96	0.0107	91.8463	7.9605	0.1479	0.0338
9/10/96	0.0108	91.8466	7.9603	0.1482	0.0335
9/11/96	0.0109	91.8452	7.9609	0.1484	0.0339
9/12/96**		91.8464	7.9599	0.1481	0.0339
9/16/96**		91.8464	7.9617	0.1467	0.0336
9/17/96**		91.8461	7.9596	0.1498	0.0329
9/18/96**		91.8466	7.9595	0.1484	0.0339
9/24/96	0.0100	91.8475	7.9597	0.1490	0.0336
9/25/96**		91.8459	7.9624	0.1474	0.0330
9/30/96	0.0108	91.8487	7.9577	0.1489	0.0339
10/1/96	0.0142	91.8435	7.9605	0.1480	0.0338
<hr/>					
Mean	0.0112	91.8455	7.9602	0.1486	0.0337
Std. Dev.	0.0017	0.0018	0.0011	0.0010	0.0003
% RSD	15.1571	0.0020	0.0139	0.6650	0.9447
Cert. (10/1/96)	0.0091	91.8529	7.9566	0.1479	0.0335
±	0.0010	0.0100	0.0100	0.0010	0.0030
<hr/>					
% Deviation	19.04684	-0.00801	0.045278	0.450705	0.539616

*Renormalized using average 238 for 7/29/96 to 9/11/96

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[illegible]

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File No. 15082

Page 2

Certified: April 13, 1995

Expires: April 12, 1997

<u>NOMINAL</u>	<u>TRUE MASS CORRECTIONS (mg)</u>	<u>APPARENT MASS CORRECTIONS (mg)</u>
100 g	+0.399	+0.163
50	+0.244	+0.126
20 (dot)	+0.016	-0.031
20	+0.064	+0.017
10	+0.076	+0.053
5	+0.054	+0.042
2 (dot)	+0.017	+0.012
2	+0.040	+0.035
1	+0.009	+0.007
500 mg	-0.018	+0.012
200 (dot)	+0.003	+0.015
200	+0.001	+0.013
100	+0.008	+0.014
50 mg	+0.009	+0.012
20 (dot)	+0.007	+0.002
20	+0.001	-0.003
10	+0.007	+0.005
5 mg	+0.002	+0.001
2 (dot)	-0.007	-0.007
2	+0.002	+0.002
1	-0.001	-0.002

M

Validation of Plutonium Oxide
PEOR3258-STD-1, As The Primary
Radiochemistry Standard

Prepared By

Nelson D. Stalnaker

Signature: Nelson D. Stalnaker Date 1/4/96

A. Introduction

This document presents the analytical methods, the analytical results and the error analysis used to validate the plutonium oxide, PEOR 3258-STD-1, as the NIST traceable primary standard for radioanalytical chemistry procedures. Traceability to NIST is established by calibrating the analytical methods with NIST standards.

B. Analytical Methods and Data

The approximately 3 grams of plutonium oxide set aside for the primary radioanalytical chemistry standard was taken from a larger batch of high purity oxide that was blended and characterized for another set of standards. Plutonium assay, plutonium isotopic, Am-241 and impurities analyses were run on the oxide. The results of the analyses are given in Table #1.

Five cuts of the oxide were analyzed for plutonium content, plutonium isotopic and Am-241 content. Three cuts were analyzed for impurities.

The plutonium assays were run by coulometry. Duplicate measurements were made on each of the five cuts giving a total of ten values. The uncertainty in the assay value is given in the analysis report as $\pm 0.1\%$ relative. These measurements were made against NBL standard CRM-126 a plutonium metal.

The plutonium isotopic were run by mass spectroscopy. A single measurement on each of the five cuts was done. These analyses are traceable to NBL standard CRM-126 and NIST standards SRM 946 ,SRM 947 and SRM 948.

The Am-241 content was run by mass spectroscopy and radiochemistry . Duplicate analyses were done on each of the five cuts. The radiochemistry results were obtained on instrumentation that is calibrated with NIST standards. The radiochemistry instrumentation was calibrated with Isotope Products Laboratories sources numbers 425-65-1 and 425-65-2 and NIST source SRM-4322. The mass spectroscopy results are not directly traceable

to NIST because appropriate standards are not available. However, the mass spectroscopy results are believed to be the more accurate results. The radiochemistry and mass spectroscopy results are in good agreement and differ by less than 1%. The activity contributed by the Am-241 to the total alpha activity is approximately 4%. Therefore, a 1% error in the Am-241 value will contribute an error of ~ 0.04% to the total activity. The mass spectroscopy results will be used in the activity evaluations.

The impurities were run by DC Arc and ICP-MS. Three cuts were analyzed.

The analytical results for all analyses are recorded under sample ID # 200016529 in the LIMS data banks.

Table # 1

Analysis Data for Primary Standard Oxide
PEOR 3258-Std-1

Assay, Wt Frxn Pu*	Am-241, ppm*	Impurities, ppm**
87.827 ± 0.042	767.9 ± 2.0 (MS) 762.6 ± 3.6 (RC)	< 5000

Plutonium Isotopic***

Isotope	Weight Fraction	%RSD
Pu 238	0.000145 ± 0.0000043	± 2.96
Pu 239	0.937614 ± 0.000006	± 0.0006
Pu 240	0.059445 ± 0.000005	± 0.0084
Pu 241	0.002237 ± 0.000002	± 0.089
Pu 242	0.000559 ± 0.000001	± 0.179

* The ± value is the standard deviation in the ten results. Five sample cuts were analyzed in duplicate.

** The detailed analyses can be found under Task ID 300038569 and 300038570 in the LIMS data bank.

*** Average and standard deviation of five analyses.

II. B. Specific Activity Evaluation

This section discusses the calculation of the gross alpha activity per gram of the plutonium oxide, which is the quantity that is used in the calibration of the counting instrumentation. The gross alpha activity of the oxide changes with time because certain isotopes decay and others grow in.

The three radionuclides that cause the greatest change in the gross alpha activity are Pu-238, Pu-241 and Am-241. The Pu-238 decays with an 87.74 year half-life to U-234 which has a half-life of 245000 years. Because of the long half-life of U-234, it does not contribute a measurable amount to the total alpha activity. The Pu-241 is a beta emitter with a 14.35 year half-life that decays mostly by beta emission to Am-241 which alpha decays with a 432 year half-life. In growth of Am-241 does contribute to the gross alpha activity. The very small amount (0.00241%) of Pu-241 that alpha decays contributes a small amount (~ 0.015%) to the gross alpha activity. The other radionuclides decay only a small amount over 10 to 20 years; however their decay will be corrected for in calculating the specific activity.

Table #2 gives the half-lives used in the gross alpha activity calculations. The half-lives are taken from references 1,2,3 and 4 and are consistent with the half-lives listed on NBS or NIST standard certificates.

The specific activity is calculated as follows using the values from Tables 1 and 2:

$$\text{Alpha Sp Act of Oxide} = (\text{Wt Frxn Pu in oxide}) ((\text{Wt Frxn Pu-238} \times \text{Sp Act of Pu-238}) + (\text{Wt Frxn Pu-239} \times \text{Sp Act of Pu-239}) + (\text{Wt Frxn Pu-240} \times \text{Sp Act of Pu-240}) + (\text{Wt Frxn Pu-241} \times \text{Sp Act of Pu-241} \times \text{Alpha Branching Ratio}) + (\text{Wt Frxn Pu-242} \times \text{Sp Act of Pu-242})) + (\text{Wt Frxn Am-241 in oxide} \times \text{Sp Act of Am-241}).$$

The alpha specific activity on 7-15-95 was 0.06773 curies per gram of oxide. Table #3 gives the specific activity over the next 10 years

II. C. Estimated Uncertainty in Total Alpha Activity

In this section an estimate of the uncertainty of the alpha specific activity is given. The alpha activity contributions of Pu-241 and Pu-242 are less than 0.015% each, and errors in these values are not considered in these calculations.

The uncertainty in activity contributed by of each of the other radionuclides is calculated as follows :

% Relative Uncertainty in the expression (Wt Frxn Pu-238 X Sp Act of Pu-238) = Sq. Root of $((2.97)^2 + (0.05)^2) = +/- 2.97$

Similarly values are calculated for Pu-239, Pu-240 and Am-241. The values are +/- 0.11, +/- 0.17 and +/- 0.51, respectively.

Next the uncertainty in the specific activity of each radionuclide is calculated as follows:

Sp Act Uncertainty due to Pu-238 = $(+/- 0.0297)(.000145)(17.1116)$
 $(0.87827) = 6.47 \text{ E-05 curies per gram of oxide.}$

Similarly values are calculated for Pu-239, Pu-240 and Am-241. The values are +/- 5.62E-05, +/- 2.01E-05 and +/- 1.17E-05, respectively

The four values from these calculations are combined as independent errors to give a total uncertainty in the specific activity due to the uncertainty in the specific activities and the isotopic composition. The uncertainty is 1.42 E-04 curies per gram of oxide. The relative uncertainty

$$= (8.87\text{E-}05)(100)/(0.06773) = +/- 0.13\%.$$

The last step in estimating the total uncertainty is to combine the uncertainty in the plutonium assay value and the above value.

$$\text{Total Uncertainty} = \text{Sq Root of } ((0.13)^2 + (0.10)^2) = +/- 0.16 \%$$

Table # 2*

Radionuclide	Half-Life, yrs**	Specific Activity, Ci/g	%RSD
Pu-238	87.74 $\pm$ 0.04 (1)	17.116 $\pm$ 0.008	$\pm$ 0.045
Pu-239	24119 $\pm$ 26(1)	0.062005 $\pm$ 0.000067	$\pm$ 0.11
Pu-240	6564 $\pm$ 11(1)	0.22696 $\pm$ 0.00038	$\pm$ 0.17
Pu-241	14.35 $\pm$ 0.02(2)	103.3 $\pm$ 0.14	$\pm$ 0.14
Pu-242	376300 $\pm$ 900(2)	0.003925 $\pm$ 0.000009	$\pm$ 0.24
Am-241	432.7 $\pm$ 0.5(3)	3.428 $\pm$ 0.004	$\pm$ 0.12

* The numbers in parenthesis are references given at the end of this document.

** The $\pm$ values are one standard deviation.

Table #3

Specific Activity as a Function of Time

Date	Specific Activity, Ci/g	Date	Specific Activity Ci/g
7/15/1995	0.06772	7/15/2000	0.06904
1/15/1996	0.06787	1/15/2001	0.06915
7/15/1996	0.06802	7/15/2001	0.06926
1/15/1997	0.06816	1/15/2002	0.06937
7/15/1997	0.06829	7/15/2002	0.06947
1/15/1998	0.06843	1/15/2003	0.06958
7/15/1998	0.06856	7/15/2003	0.06967
1/15/1999	0.06868	1/15/2004	0.06977
7/15/1999	0.06880	7/15/2004	0.06987
1/15/2000	0.06893	1/15/2005	0.06995

References:

1. Strohn, Walter, Int. J. Appl. Radiat. Isot. Vol 29, No. 8 pp. 481-483, 1979.
2. Strohn, Walter, Int. J. Appl. Radiat. Isot. Vol 35, No.3, p155-'57, 1984.
3. Reilly, Doug et. al., "Passivs Nondestructive Assay of Nuclear Materials" NUREG/CR-5550, LA-UR-90-732, Published March 1991.
4. Browne, Edgardo et. al. "Table of Radioactive Isotopes" Published by Wiley & Sons,Inc 1986.

Los Alamos
NATIONAL LABORATORY
memorandum

Technology and Safety Assessment Division
TSA-1 • Statistics Group

To/MS: Robert Marshall, CST-8, MS G740
From/MS: L. Bruckner & L. Ticknor, TSA-1, F600
Phone/Fax: 7-6246 / Fax 7-4470
email: larry@lanl.gov, lot@lanl.gov
Symbol: TSA-1:96-117
Date: April 23, 1996

Uncertainties For the Radiochemistry and PDP Standards

We have analyzed the data received from you and Nelson Stalnaker on this material. The data was provided in the report, "Validation of Plutonium Oxide PEOR3258-STD-1 as the Primary Radiochemistry Standard" Stalnaker, January 4, 1996, in two Excel spreadsheets, and in supplementary notes. The first document contained the basic information on the material itself. The other sources contained information on 63 standards made for the oxide. We were asked to provide uncertainty estimates for a number of parameters relating to this material and the standards. The parameters were: the specific alpha activity of the oxide and, for each standard, the total alpha activity, the plutonium content, and the americium content.

UNCERTAINTIES. For each parameter of interest we computed a 95% confidence interval (or 95% confidence bounds) for the true value of the parameter. To do so, we applied propagation of variance (POV) techniques to the calculational formula for the parameter. The POV technique involves appropriate combining of the uncertainties of all the components in the formula. Where we did not have well-supported estimates on uncertainties, we took a conservative approach and used values that we believed exceeded the actual values. The 95% confidence intervals (bounds) presented are believed to be larger than they would be if better estimates were available. Thus, the expressions "at least 95%" confidence interval" or "at-least" 95% confidence bounds.

HOMOGENEITY. Five samples were selected and analyzed by coulometry for fraction plutonium. The sample results show that the Pu fractions are slightly higher for samples 1 and 2 than for samples 3, 4 and 5. The average fraction Pu of the first 2 samples is 0.0006 greater than the average for the last 3 samples. This difference might be due to chance or to a real difference in chemical procedure or in the oxide. While the difference is not unusual for oxides, the fact that it occurs on the first two samples is interesting. If there is any concern, it would be useful to know the locations in the oxide from which the samples were selected, the sample selection procedure, details on the sample preparation procedure, and the date, time, operator and reagents used in the analyses.

ALPHA SPECIFIC ACTIVITY. The specific activity (SA) was calculated in Stalnaker's report to be 0.06773 curies/gram oxide. We calculated 95% confidence intervals for the SA value under two sets of conditions.

1. The isotopic specific activity values were considered to have uncertainties associated with them. The uncertainties are given by Stalnaker in terms of percent relative standard devi-

ations.

2. The uncertainties on isotopic specific activity values were ignored.

For the first case an at-least 95% confidence interval for the true SA value is $0.06773 \pm 0.36\%$, while for the second case the at-least 95% confidence interval is $0.06773 \pm 0.32\%$.

ALPHA ACTIVITY. The activity, A, for an item is obtained from the formula $A = SA * W$ where W is the weight of the oxide in grams. The uncertainty associated with A is then a combination of the uncertainty associated with SA and the uncertainty associated with W. The values of SA and W directly enter into the formula. Thus, each of the 63 items will have its own activity value and uncertainty. The standards are in 3 groupings, 30 mg, 300mg, and 3000 mg of oxide. Within each grouping the activity and uncertainties are essentially the same. Hence the results for the alpha activity can be summarized in terms of weight class.

Pu	PuO ₂	Specific Activity	Activity	At-Least 95% Confidence Bound
mg	mg	curies/g	curies	%
30	34.16	0.06773	2.314E-03	0.39
300	341.58	0.06773	2.314E-02	0.36
3000	3415.81	0.06773	2.314E-01	0.36

Thus, for the 300 mg standards, a 95% confidence interval for the alpha activity is $0.02314 \pm 0.36 * 0.02314 / 100$ or 0.02314 ± 0.00008 curies.

Pu AND AMERICIUM CONTENT. The PuO₂ mass and americium loss values were provided in the Excel spreadsheets. We computed uncertainties for for the plutonium and americium content of each standard considering both random and systematic effects. The attached table gives the "at-least" 95% confidence bounds for each standard. The bounds are expressed as percents of the plutonium and americium contents respectively.

Attachment: Tables giving the individual uncertainties for the 63 standards.

Distribution:

N. Stalnaker, CST-8, MS G740

Authors

TSA-1 file

N

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO_2 -Diatomaceous Earth Standard LLWPU0.1G

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	0.11589 grams	± 0.00008 grams ^(a)
Pu Assay	87.897 % of PuO_2 *	± 0.081 % ^(b)
Pu in standard	0.10186 grams	± 0.00014 grams ^(c)
^{241}Am Assay	1140.3 $\mu\text{g/g}$ PuO_2 *	± 0.3 $\mu\text{g/g}$ ^(d)
^{241}Am in standard	0.1321 mg*	± 0.0009 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	8.138 mCi**	± 0.023 mCi ^(g)
	3.011E+08 Bq	$\pm 9\text{E}+05$ Bq ^(g)

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

Bill J. McKerley, Group Leader
CMR Analytical Chemistry

Robert S. Marshall, Project Manager
NDA WRM Production

MEASUREMENT METHOD AND TRACEABILITY

The nuclear contents of this standard were characterized and quantified using the following methods with traceability to the National Institute of Standards and Technology (NIST) or New Brunswick Laboratory (NBL) standards.

<u>Measurement</u>	<u>Measurement Method</u>	<u>Reference Material</u>
Weighing	5 place analytical balance	NIST traceable weights
Pu Assay	Controlled Potential Coulometry	NBL CRM # 126
Am Assay	Isotope Dilution Mass Spectrometry	NBL CRM # 128
Pu Isotopic	Total Evaporation Mass Spectrometry	NBL CRM # 128
Alpha Activity	Calculated from Pu and Am mass, and isotope alpha specific activities listed below.	Pu $t_{1/2}$ as stated for NIST SRM946 Am $t_{1/2}$ as stated for NIST SRM4322B

The stated Alpha Activity was calculated using the following isotope half lives (in years): ^{238}Pu : 87.74; ^{239}Pu : 24,119; ^{240}Pu : 6,560; ^{241}Pu : 14.34; ^{242}Pu : 387,000; and ^{241}Am : 432.2.

NOTES

Random and systematic error terms were combined and reported as 95% confidence intervals. Error terms include estimates of the following:

- (a) Balance precision, buoyancy, standard weight bias, and transfer loss.
- (b) Long and short term precision, control check bias, weighing precision and bias.
- (c) Above weighing and assay terms, and transfer loss.
- (d) Long and short term precision, and bias.
- (e) Above weighing terms, Am assay terms, and transfer loss.
- (f) Long and short term precision, and bias.
- (g) Based on propagated uncertainties on the Pu mass, ^{241}Am mass and Pu isotopic uncertainties listed above. No half-life uncertainty components are included and it is recommended that all facilities participating in the NDA PDP program use the half-lives listed above to preclude facility-to-facility error terms introduced by using different half life values.

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard LLWPU1.0G

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	1.13506 grams	± 0.00010 grams ^(a)
Pu Assay	87.897 % of PuO ₂ *	± 0.081 % ^(b)
Pu in standard	0.99769 grams	± 0.00098 grams ^(c)
²⁴¹ Am Assay	1140.3 µg/g PuO ₂ *	± 0.3 µg/g ^(d)
²⁴¹ Am in standard	1.2940 mg *	± 0.0086 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	79.71 mCi ** 2.949E+09 Bq	± 0.23 mCi ^(g) ± 9E+06 Bq ^(g)

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

Bill J. McKerley, Group Leader
CMR Analytical Chemistry

Robert S. Marshall, Project Manager
NDA WRM Production

MEASUREMENT METHOD AND TRACEABILITY

The nuclear contents of this standard were characterized and quantified using the following methods with traceability to the National Institute of Standards and Technology (NIST) or New Brunswick Laboratory (NBL) standards.

<u>Measurement</u>	<u>Measurement Method</u>	<u>Reference Material</u>
Weighing	5 place analytical balance	NIST traceable weights
Pu Assay	Controlled Potential Coulometry	NBL CRM # 126
Am Assay	Isotope Dilution Mass Spectrometry	NBL CRM # 128
Pu Isotopic	Total Evaporation Mass Spectrometry	NBL CRM # 128
Alpha Activity	Calculated from Pu and Am mass, and isotope alpha specific activities listed below.	Pu $t_{1/2}$ as stated for NIST SRM946 Am $t_{1/2}$ as stated for NIST SRM4322B

The stated Alpha Activity was calculated using the following isotope half lives (in years): ^{238}Pu : 87.74; ^{239}Pu : 24,119; ^{240}Pu : 6,560; ^{241}Pu : 14.34; ^{242}Pu : 387,000; and ^{241}Am : 432.2.

NOTES

Random and systematic error terms were combined and reported as 95% confidence intervals. Error terms include estimates of the following:

- (a) Balance precision, buoyancy, standard weight bias, and transfer loss.
- (b) Long and short term precision, control check bias, weighing precision and bias.
- (c) Above weighing and assay terms, and transfer loss.
- (d) Long and short term precision, and bias.
- (e) Above weighing terms, Am assay terms, and transfer loss.
- (f) Long and short term precision, and bias.
- (g) Based on propagated uncertainties on the Pu mass, ^{241}Am mass and Pu isotopic uncertainties listed above. No half-life uncertainty components are included and it is recommended that all facilities participating in the NDA PDP program use the half-lives listed above to preclude facility-to-facility error terms introduced by using different half life values.

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard LLWPU10.0G

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	11.3739 grams	± 0.00033 grams ^(a)
Pu Assay	87.897 % of PuO ₂ *	± 0.081 % ^(b)
Pu in standard	9.99731 grams	± 0.00938 grams ^(c)
²⁴¹ Am Assay	1140.3 µg/g PuO ₂ *	± 0.3 µg/g ^(d)
²⁴¹ Am in standard	12.967 mg*	± 0.086 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	798.8 mCi** 2.955E+10 Bq	± 2.3 mCi ^(g) ± 8E+07 Bq ^(g)

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

 Bill J. McKerley, Group Leader
 CMR Analytical Chemistry

 Robert S. Marshall, Project Manager
 NDA WRM Production

MEASUREMENT METHOD AND TRACEABILITY

The nuclear contents of this standard were characterized and quantified using the following methods with traceability to the National Institute of Standards and Technology (NIST) or New Brunswick Laboratory (NBL) standards.

<u>Measurement</u>	<u>Measurement Method</u>	<u>Reference Material</u>
Weighing	5 place analytical balance	NIST traceable weights
Pu Assay	Controlled Potential Coulometry	NBL CRM # 126
Am Assay	Isotope Dilution Mass Spectrometry	NBL CRM # 128
Pu Isotopic	Total Evaporation Mass Spectrometry	NBL CRM # 128
Alpha Activity	Calculated from Pu and Am mass, and isotope alpha specific activities listed below.	Pu $t_{1/2}$ as stated for NIST SRM946 Am $t_{1/2}$ as stated for NIST SRM4322B

The stated Alpha Activity was calculated using the following isotope half lives (in years): ^{238}Pu : 87.74; ^{239}Pu : 24,119; ^{240}Pu : 6,560; ^{241}Pu : 14.34; ^{242}Pu : 387,000; and ^{241}Am : 432.2.

NOTES

Random and systematic error terms were combined and reported as 95% confidence intervals. Error terms include estimates of the following:

- (a) Balance precision, buoyancy, standard weight bias, and transfer loss.
- (b) Long and short term precision, control check bias, weighing precision and bias.
- (c) Above weighing and assay terms, and transfer loss.
- (d) Long and short term precision, and bias.
- (e) Above weighing terms, Am assay terms, and transfer loss.
- (f) Long and short term precision, and bias.
- (g) Based on propagated uncertainties on the Pu mass, ^{241}Am mass and Pu isotopic uncertainties listed above. No half-life uncertainty components are included and it is recommended that all facilities participating in the NDA PDP program use the half-lives listed above to preclude facility-to-facility error terms introduced by using different half life values.

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard LLWPU25.0G

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	28.4388 grams	± 0.00063 grams ^(a)
Pu Assay	87.897 % of PuO ₂ *	± 0.081 % ^(b)
Pu in standard	24.9969 grams	± 0.0234 grams ^(c)
²⁴¹ Am Assay	1140.3 µg/g PuO ₂ *	± 0.3 µg/g ^(d)
²⁴¹ Am in standard	32.421 mg *	± 0.215 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		
	1997 mCi **	± 6 mCi ^(g)
	7.390E+10 Bq	± 2E+08 Bq ^(g)

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

 Bill J. McKerley, Group Leader
 CMR Analytical Chemistry

 Robert S. Marshall, Project Manager
 NDA WRM Production

MEASUREMENT METHOD AND TRACEABILITY

The nuclear contents of this standard were characterized and quantified using the following methods with traceability to the National Institute of Standards and Technology (NIST) or New Brunswick Laboratory (NBL) standards.

<u>Measurement</u>	<u>Measurement Method</u>	<u>Reference Material</u>
Weighing	5 place analytical balance	NIST traceable weights
Pu Assay	Controlled Potential Coulometry	NBL CRM # 126
Am Assay	Isotope Dilution Mass Spectrometry	NBL CRM # 128
Pu Isotopic	Total Evaporation Mass Spectrometry	NBL CRM # 128
Alpha Activity	Calculated from Pu and Am mass, and isotope alpha specific activities listed below.	Pu $t_{1/2}$ as stated for NIST SRM946 Am $t_{1/2}$ as stated for NIST SRM4322B

The stated Alpha Activity was calculated using the following isotope half lives (in years): ^{238}Pu : 87.74; ^{239}Pu : 24,119; ^{240}Pu : 6,560; ^{241}Pu : 14.34; ^{242}Pu : 387,000; and ^{241}Am : 432.2.

NOTES

Random and systematic error terms were combined and reported as 95% confidence intervals. Error terms include estimates of the following:

- Balance precision, buoyancy, standard weight bias, and transfer loss.
- Long and short term precision, control check bias, weighing precision and bias.
- Above weighing and assay terms, and transfer loss.
- Long and short term precision, and bias.
- Above weighing terms, Am assay terms, and transfer loss.
- Long and short term precision, and bias.
- Based on propagated uncertainties on the Pu mass, ^{241}Am mass and Pu isotopic uncertainties listed above. No half-life uncertainty components are included and it is recommended that all facilities participating in the NDA PDP program use the half-lives listed above to preclude facility-to-facility error terms introduced by using different half life values.

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard LLWPU65.0G

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	73.9437 grams	± 0.00118 grams ^(a)
Pu Assay	87.897 % of PuO ₂ *	± 0.081 % ^(b)
Pu in standard	64.9943 grams	± 0.0606 grams ^(c)
²⁴¹ Am Assay	1140.3 µg/g PuO ₂ *	± 0.3 µg/g ^(d)
²⁴¹ Am in standard	84.298 mg *	± 0.559 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		
	5193 mCi **	± 15 mCi ^(g)
	1.921E+11 Bq	± 6E+08 Bq ^(g)

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

 Bill J. McKerley, Group Leader
 CMR Analytical Chemistry

 Robert S. Marshall, Project Manager
 NDA WRM Production

MEASUREMENT METHOD AND TRACEABILITY

The nuclear contents of this standard were characterized and quantified using the following methods with traceability to the National Institute of Standards and Technology (NIST) or New Brunswick Laboratory (NBL) standards.

<u>Measurement</u>	<u>Measurement Method</u>	<u>Reference Material</u>
Weighing	5 place analytical balance	NIST traceable weights
Pu Assay	Controlled Potential Coulometry	NBL CRM # 126
Am Assay	Isotope Dilution Mass Spectrometry	NBL CRM # 128
Pu Isotopic	Total Evaporation Mass Spectrometry	NBL CRM # 128
Alpha Activity	Calculated from Pu and Am mass, and isotope alpha specific activities listed below.	Pu $t_{1/2}$ as stated for NIST SRM946 Am $t_{1/2}$ as stated for NIST SRM4322B

The stated Alpha Activity was calculated using the following isotope half lives (in years): ^{238}Pu : 87.74; ^{239}Pu : 24,119; ^{240}Pu : 6,560; ^{241}Pu : 14.34; ^{242}Pu : 387,000; and ^{241}Am : 432.2.

NOTES

Random and systematic error terms were combined and reported as 95% confidence intervals. Error terms include estimates of the following:

- (a) Balance precision, buoyancy, standard weight bias, and transfer loss.
- (b) Long and short term precision, control check bias, weighing precision and bias.
- (c) Above weighing and assay terms, and transfer loss.
- (d) Long and short term precision, and bias.
- (e) Above weighing terms, Am assay terms, and transfer loss.
- (f) Long and short term precision, and bias.
- (g) Based on propagated uncertainties on the Pu mass, ^{241}Am mass and Pu isotopic uncertainties listed above. No half-life uncertainty components are included and it is recommended that all facilities participating in the NDA PDP program use the half-lives listed above to preclude facility-to-facility error terms introduced by using different half life values.

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard LLWPU67.0G

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	76.1905 grams	± 0.00147 grams ^(a)
Pu Assay	87.897 % of PuO ₂ *	± 0.081 % ^(a)
Pu in standard	66.9691 grams	± 0.0626 grams ^(a)
²⁴¹ Am Assay	1140.3 µg/g PuO ₂ *	± 0.3 µg/g ^(a)
²⁴¹ Am in standard	86.859 mg*	± 0.576 mg ^(a)
Isotopic Weight Fraction		
²³⁸ Pu	0.000149	± 0.000005 ^(a)
²³⁹ Pu	0.929845	± 0.000015 ^(a)
²⁴⁰ Pu	0.067206	± 0.000007 ^(a)
²⁴¹ Pu	0.002141*	± 0.000009 ^(a)
²⁴² Pu	0.000659	± 0.000003 ^(a)
Total Alpha Activity in standard		
	5351 mCi**	± 15 mCi ^(a)
	1.980E+11 Bq	± 6E+08 Bq ^(a)

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

 Bill J. McKerley, Group Leader
 CMR Analytical Chemistry

 Robert S. Marshall, Project Manager
 NDA WRM Production

MEASUREMENT METHOD AND TRACEABILITY

The nuclear contents of this standard were characterized and quantified using the following methods with traceability to the National Institute of Standards and Technology (NIST) or New Brunswick Laboratory (NBL) standards.

<u>Measurement</u>	<u>Measurement Method</u>	<u>Reference Material</u>
Weighing	5 place analytical balance	NIST traceable weights
Pu Assay	Controlled Potential Coulometry	NBL CRM # 126
Am Assay	Isotope Dilution Mass Spectrometry	NBL CRM # 128
Pu Isotopic	Total Evaporation Mass Spectrometry	NBL CRM # 128
Alpha Activity	Calculated from Pu and Am mass, and isotope alpha specific activities listed below.	Pu $t_{1/2}$ as stated for NIST SRM946 Am $t_{1/2}$ as stated for NIST SRM4322B

The stated Alpha Activity was calculated using the following isotope half lives (in years): ^{238}Pu : 87.74; ^{239}Pu : 24,119; ^{240}Pu : 6,560; ^{241}Pu : 14.34; ^{242}Pu : 387,000; and ^{241}Am : 432.2.

NOTES

Random and systematic error terms were combined and reported as 95% confidence intervals. Error terms include estimates of the following:

- Balance precision, buoyancy, standard weight bias, and transfer loss.
- Long and short term precision, control check bias, weighing precision and bias.
- Above weighing and assay terms, and transfer loss.
- Long and short term precision, and bias.
- Above weighing terms, Am assay terms, and transfer loss.
- Long and short term precision, and bias.
- Based on propagated uncertainties on the Pu mass, ^{241}Am mass and Pu isotopic uncertainties listed above. No half-life uncertainty components are included and it is recommended that all facilities participating in the NDA PDP program use the half-lives listed above to preclude facility-to-facility error terms introduced by using different half life values.

Los Alamos National Laboratory

Certificate of Content and Traceability

PuO₂-Diatomaceous Earth Standard LLWPU68.0G

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	77.3434 grams	± 0.00133 grams ^(a)
Pu Assay	87.897 % of PuO ₂ *	± 0.081 % ^(b)
Pu in standard	67.9826 grams	± 0.0634 grams ^(c)
²⁴¹ Am Assay	1140.3 µg/g PuO ₂ *	± 0.3 µg/g ^(d)
²⁴¹ Am in standard	88.173 mg*	± 0.584 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		
	5432 mCi**	± 16 mCi ^(g)
	2.010E+11 Bq	± 6E+08 Bq ^(g)

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

 Bill J. McKerley, Group Leader
 CMR Analytical Chemistry

 Robert S. Marshall, Project Manager
 NDA WRM Production

MEASUREMENT METHOD AND TRACEABILITY

The nuclear contents of this standard were characterized and quantified using the following methods with traceability to the National Institute of Standards and Technology (NIST) or New Brunswick Laboratory (NBL) standards.

<u>Measurement</u>	<u>Measurement Method</u>	<u>Reference Material</u>
Weighing	5 place analytical balance	NIST traceable weights
Pu Assay	Controlled Potential Coulometry	NBL CRM # 126
Am Assay	Isotope Dilution Mass Spectrometry	NBL CRM # 128
Pu Isotopic	Total Evaporation Mass Spectrometry	NBL CRM # 128
Alpha Activity	Calculated from Pu and Am mass, and isotope alpha specific activities listed below.	Pu $t_{1/2}$ as stated for NIST SRM946 Am $t_{1/2}$ as stated for NIST SRM4322B

The stated Alpha Activity was calculated using the following isotope half lives (in years): ^{238}Pu : 87.74; ^{239}Pu : 24,119; ^{240}Pu : 6,560; ^{241}Pu : 14.34; ^{242}Pu : 387,000; and ^{241}Am : 432.2.

NOTES

Random and systematic error terms were combined and reported as 95% confidence intervals. Error terms include estimates of the following:

- (a) Balance precision, buoyancy, standard weight bias, and transfer loss.
- (b) Long and short term precision, control check bias, weighing precision and bias.
- (c) Above weighing and assay terms, and transfer loss.
- (d) Long and short term precision, and bias.
- (e) Above weighing terms, Am assay terms, and transfer loss.
- (f) Long and short term precision, and bias.
- (g) Based on propagated uncertainties on the Pu mass, ^{241}Am mass and Pu isotopic uncertainties listed above. No half-life uncertainty components are included and it is recommended that all facilities participating in the NDA PDP program use the half-lives listed above to preclude facility-to-facility error terms introduced by using different half life values.