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DESIGN AND FABRICATION OF NDA STANDARDS*

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ABSTRACT

Standards are needed to calibrate various NDA (Non-destructive Assay) instruments such as neutron coincidence counters, gamma-ray counters, and calorimeters. These instruments measure various ranges of nuclear material being produced in the DOE nuclear community. Los Alamos National Laboratory has taken a lead role in fabrication of uranium and plutonium standards, along with other actinides such as neptunium and americium. These standards have been fabricated for several laboratories within the complex. This paper will summarize previous publications detailing the careful planning encompassing components such as precise weighing, destructive analysis, and the use of post fabrication NDA measurements to confirm that the standards meet all preliminary expectations before use in instrument calibration. The paper will also describe the specialized containers, diluents, and the various amount of nuclear materials needed to accommodate the calibration ranges of the instruments.

INTRODUCTION

The Plutonium Facility, TA-55, at Los Alamos National Laboratory is currently producing NDA calibration standards used by various laboratories in the DOE complex. These NIST traceable standards have been produced to calibrate NDA instruments for accountability measurements used for resolving shipper/receiver differences, and for accountability in process residues and process waste. Standards have been fabricated from various isotopes of plutonium or uranium, including other actinides such as neptunium and americium. This paper will summarize the meticulous planning that is needed for fabricating standards.

STANDARD PREPARATION

Containers

Containerization is a critical step in standards fabrication work. Each standard task is carefully analyzed during the planning stage to design and construct a container that is easily handled in the glovebox environment and compatible with the instrument it was designed for. Specialized containers have been designed for both SGS, neutron, and calorimeters.

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Two types of Segmented Gamma Scanners exist at TA-55. The first is a helical can scanner with a typical collimator height of 12.5 inches. The ideal container for SGS measurements would be a tall, cylindrical container approximately 100 mm in diameter and 250 mm high. Specialized stainless steel cans were designed to fit the criteria for the small SGS. An inner and outer can were designed and fabricated (See Figure 1). The sides of the can were cut out of 0.4 mm thick stainless steel sheets and laser welded. The 0.6 mm tops and bottoms were stamped out of the sheets with the bottom cap laser welded. The height of the inner can was 272 mm and the diameter was 95.4 mm. The height and diameter of the outer can are slightly larger.

The second type of gamma scanner is the large helical SGS. This instrument is used to measure material typically packaged in 30 or 55 gallon drums, although it can measure items in smaller can configurations such as 5 gallon canisters. The drum standard consists of a modified 55 gallon drum (See Figure 2). The drum is loaded with twenty, four liter polyethylene bottles, each containing a quantitative amount of oxide and diluted with diatomaceous earth. A set of plutonium and uranium drum standards were fabricated ranging from 30 to 200 grams. The loadings of the each of the polyethylene bottles will be discussed later in the paper.

Figure 1

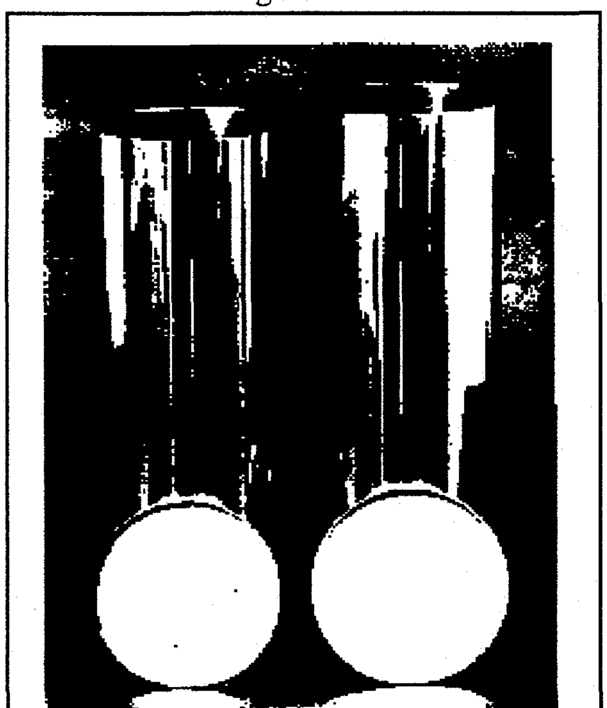
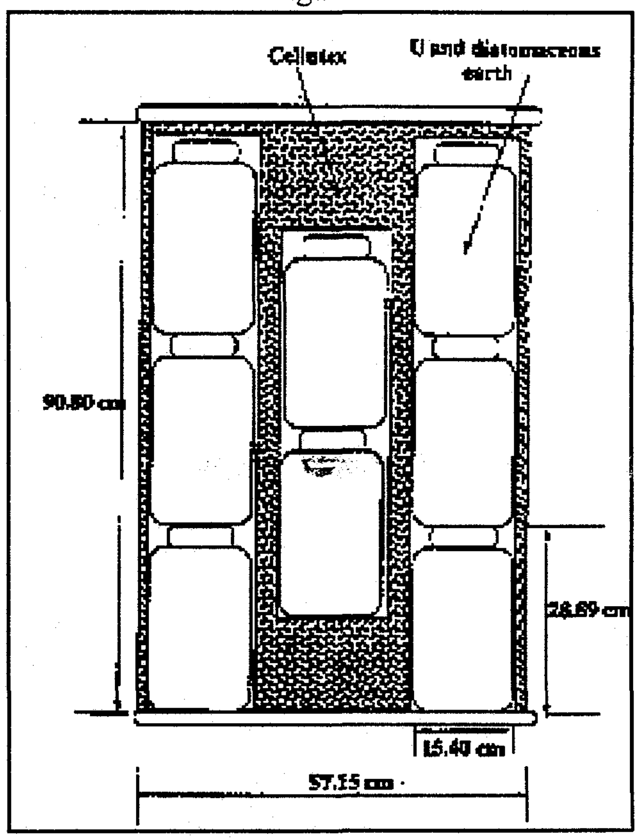


Figure 2



Small neutron coincidence counters can also use the same SGS standards for neutron calibration. Also available are mechanically sealed, doubly contained, food packaging cans loaded with undiluted oxide. For the large neutron drum counter (Shuffler), a drum standard was designed so the actinide loadings could be manipulated manually to achieve the low to high ends of calibration. The drum is filled with cellutex layers and cored to fit aluminum tubes (See Figure 3). The drum is loaded with plutonium or uranium zircalloy vials to match the needed calibration range. The zircalloy vials have a 1/16" wall thickness to minimize self absorption. For handling and safety reasons, the plutonium vials were doubly encapsulated. The uranium oxide was singly encapsulated in the outer vials. The outer vials are five inches long with a diameter of one inch. The inner vial is 3/4" in diameter and a nominal size smaller than the five-inch long vial (See Figure 4). Vial spacers filled only with diatomaceous earth was also fabricated. They allow for separation of the oxide vials and to eliminate voids in the aluminum tubing when loading the drum.

Figure 3

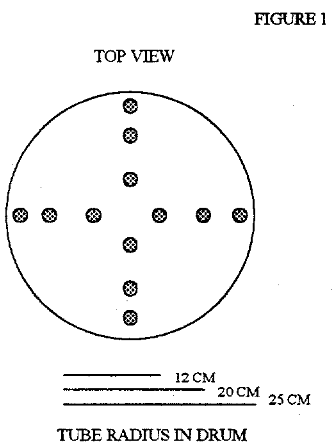
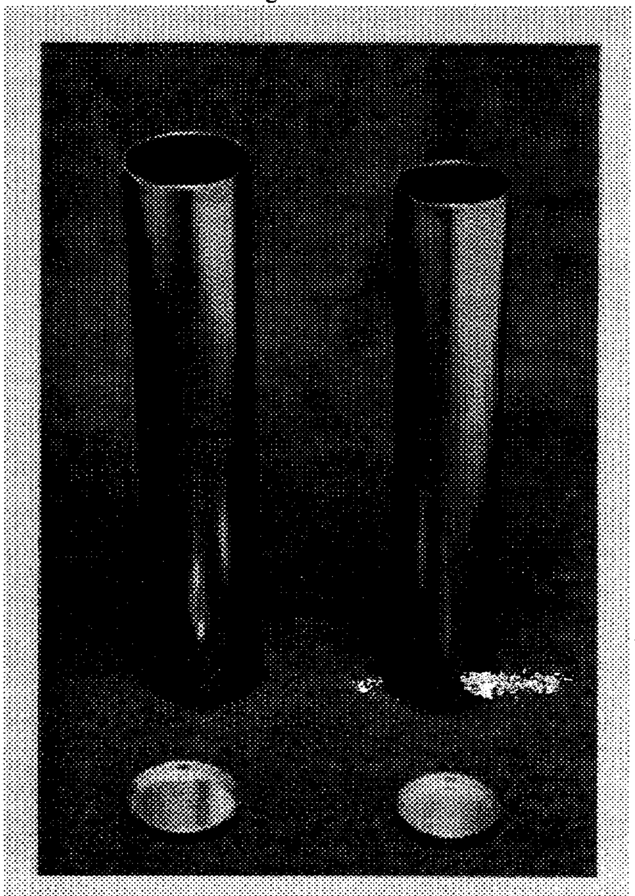


Figure 4



Matrix

Diatomaceous earth was used as a diluent in the vials. The particle size of the diatomaceous earth is between 75 μm to 106 μm with the density at 0.26.[2] Previous studies using graphite as a diluent showed a greater attenuation of the neutrons during measurements than that of diatomaceous earth. The use of diatomaceous earth in other previous experimental standards has tended us to lean towards it over graphite. Further studies and measurements in the diatomaceous earth standards display that the actinide shows little separation or segregation from the diatomaceous earth over long periods of time. Ordinary handling by the technicians during measurements has ensured occasional mixing and has allowed the actinide to continue to be homogeneous with the diatomaceous earth.

Oxide Preparation

Both the plutonium and uranium oxide are high-fired at 950° C. The materials were blended for approximately four hours in a V-type blender and sieved through a 100-mesh screen (<150μm). Samples were then taken at various areas of the blend and analyzed by the analytical group, CST-1 of the Los Alamos National Laboratory. The plutonium and uranium oxide were used in the SGS can standards, PAN standards, SGS drum standards, and the Shuffler standards. See Table I for examples of previous analytical results.

Table 1. Plutonium and Uranium Analytical Results

Plutonium Analytical

SAMPLE ID		CLS-1#	PU238	PU239	PU240	PU241	PU242	AM241(PPM)	LOI%	PU RECEIVED %	PU CALCINED %
PEOM77S1		75885	0.0125	93.735	5.993	0.2261	0.0336	118	0.11	87.84	87.97
PEOM77S2		75886	0.013	93.742	5.998	0.2235	0.0333	115	0.13	87.93	88.06
PEOM77S3		75887	0.0128	93.732	5.995	0.2244	0.035	117	0.08	87.94	88.05
PEOM77S4		75888	0.0127	93.734	5.993	0.2268	0.0331	117	0.04	87.88	87.86
PEOM77S5		75889	0.0124	93.745	5.984	0.2249	0.0338	117	0.06	87.94	87.93
	AVERAGE		0.01268000	93.73760000	5.99260000	0.22514000	0.03376000	116.80000000	0.08400000	87.90600000	87.97400000
	STDEV		0.00023875	0.00559464	0.00522494	0.00132023	0.00074364	1.09544512	0.03646917	0.04449719	0.08384510
	RSD (%)		1.882860629	0.005968406	0.087189871	0.586402795	2.202724223	0.937881092	43.41567269	0.05061906	0.095306676

Uranium Analytical

Table I. Cont.				
Sample ID	**Uranium (Wt %)**	**^{235}U (At.%)**	**^{235}U (Wt.%)**	**LOI (%)**
78571	84.34	92.18	92.11	0.01
78572	84.29	92.18	92.11	0.04
78573	84.35	92.18	92.11	0.01
78574	84.29	92.20	92.12	0.08
78575	84.38	92.18	92.11	0.06
Average	84.33	92.18	92.11	
STD	0.039	0.009	0.005	
RSD	0.05	0.01	0.01	

Actinide Loadings

The actinide loadings and procedures for handling each container differ for each set of standards fabricated. Special precautions are needed when handling the containers in the glovebox environment. To minimize the contamination of the inner containers, each can, vial, or bottle was weighed and then wrapped with plasticand or foil before introduction into the glovebox line. After introducing the container, it is tared again. This will be our "working tare weight". A calculated aliquot of actinide material was carefully measured into the container, reweighed, and documented. For the standards that are to be diluted, diatomaceous earth is added. The earth is added to the container until it is approximately 70 to 90 percent full. A void was needed to facilitate the blending of the oxide with the diatomaceous earth. The containers were then reweighed and documented. The undiluted standards were filled with the necessary amount of oxide and weighed. See tables II, III, and IV for examples of the loadings used in some of the various standards fabricated.

Table II. Shuffler Drum Standard Vials

Plutonium Vials				Uranium Vials		
# of vials	Unit mass (grams)	Total mass (grams)		# of vials	Unit mass (grams)	Total mass (grams)
55	4	220		50	4	200
4	2	8		4	2	8
6	1	6		4	1	4
4	0.5	2		4	0.5	2

Table III. Plutonium SGS Drum Standards

Number	Sample ID	SNM(Pu)	SNM/Bottle
1	STDSGPD1	30g	1.5g
1	STDSGPD2	100g	5.0g
1	STDSGPD3	200g	10.0

Table IV. SGS Can Standards

Item ID	Pu-239 (grams)		Item ID	Pu-239(grams)
STDSGMC-1	48.142		STDSGB10	9.624
STDSGMC-2	48.13		STDSGB30	28.89
STDSGMC-3	48.131		STDSGB100	96.273
STDSGMC-4	48.132		STDSGB250	192.528
STDSGMC-5	48.139		STDSGC10	9.625
STDSGMC-6	48.13		STDSGC30	28.876
STDSGA10	9.629		STDSGC100	96.268
STDSGA30	28.874		STDSGC250	240.664
STDSGA100	96.269		STDSGCAL20	19.271
STDSGA250	240.661		STDSGCAL200	192.527

Welding or Sealing of Containers

The SGS can standards, PAN standards, Np standards, and Shuffler standards were packaged in specialized container that had to be TIG (Automated Tungsten Inert Gas) welded. The plastic and or foil covered can or vials were taken to a specialized helium filled, welding glovebox and stripped of their coverings. The containers were then placed into a copper heat sink, fitted with their caps and welded. The containers are then The plutonium and uranium loaded vials and cans were helium leaked tested (10^{-7} cc/sec), transferred from the glovebox line and manually decontaminated in an open faced hood. and placed into the outer vial or can. These were placed into a bell welding jar and welded. Final weights were taken and documented.

The SGS drum standard consisted of four liter polyethylene bottles with quantitative amounts of actinide. The bottles were also plastic wrapped when introduced into the glovebox. After loading, the bottles were withdrawn from the glovbox via a bagout plastic bag. The primary bag was placed into a secondary bag, heat sealed and taped. Each bottle was checked for contamination and then strategically placed into the drum.

Blending

All of the SGS standards were blended in a V-type blender. The cans and bottles were tumbled for approximately eight to ten hours. They were then hand checked by a portable gamma monitor to check for homogeneity. If the readings demonstrated non-homogeneity, the bottle or can were placed back into the blender and remixed. The Shuffler vials were mixed in a new blender called a Turbula blender. The Turbula blender uses and X,Y, Z mixing motion with a variety of speeds. The V-type blender uses an X, Y motion with only one speed available for mixing. Mixing times were shortened to thirty minutes versus the eight to ten hours of the V-type blender. The vials were gamma monitored and reblended if needed. Recently, new SGS standards have been fabricated with only a ½ inch void for blending. We achieved homogeneity by mixing for three hours. Segmented studies by SGS show that the larger the amount of actinide added, the easier it is to achieve a homogeneous mixture at lower mixing times. Anything below 30 grams seems to need a few hours longer.

Verification

Uniformity distribution testing was done on all the SGS standards by measuring 1/4 inch segments on the SGS. The data on the vertical scan was analyzed and plotted. For the neutron Shuffler standards, the vials were also checked for homogeneity by SGS. This was to assure that the actinide was not lumping or becoming stratified in the vials. Figure 5 show a plot of the segmented measurement done on STDSGA-100 (a 100 grams standard) of the SGS containers.

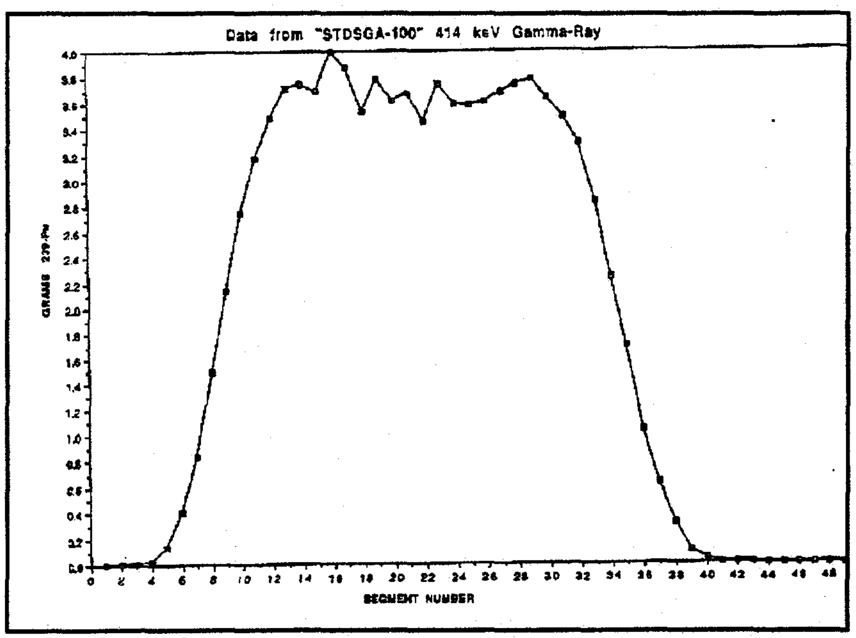


Figure 5

CONCLUSION

"Calibration is only as good as your standards." Superior actinide standards are crucial to providing good measurements of special nuclear materials. Without good standards, the NDA complex would be experiencing large inventory differences due to poor calibration of NDA instrumentation. Los Alamos National Laboratory has taken a large step into providing the NDA realm with good quality calibration standards.

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