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DESIGN AND FABRICATION OF SGS PLUTONIUM STANDARDS*

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ABSTRACT

This paper describes our experience of fabricating four sets of plutonium segmented gamma scanner (SGS) can standards. The fabrication involves careful planning, meticulous execution in weighing the plutonium oxide while minimizing contamination, chemical analyses by three different national laboratories to get accurate and independent plutonium concentrations, vertical scanning to assure mixing of the plutonium and the diluent, and finally the nondestructive verification measurement. By following these steps, we successfully fabricated 4 sets or 20 SGS can standards.

INTRODUCTION

The segmented gamma scanner (SGS) has become an important instrument for assaying the special nuclear material (SNM) content in low-density scrap and waste. To perform these assays, standards are necessary to calibrate the response of the system, which includes the detector efficiency, the amount of absorption in front of the detector, and the collimator geometry. We followed the American National Standards Institute (ANSI) standard, which provided the guidelines for the general preparation of SGS standards.[1] This paper discusses the actual design and fabrication experience gained from the can standards, which resulted in several sets of successful SGS standards. The fabrication of these standards was discussed at another conference.[2]

The standards have to satisfy the requirements of the measurement principles of the SGS assay technique; it is not necessary to duplicate the chemical form or composition of the unknown samples. The requirements for SGS standards are summarized below:

1. The SNM should be uniformly distributed in the standards.
2. The standard should have a diameter so that the gamma-ray transmission through the standard is reasonable ($0.1 < T < 0.6$).
3. The height of the standard containing SNM should be at least ten times the height of the collimator used in the SGS measurement.
4. The particle size of the SNM should be small so that the self-absorption of the particle is negligible.

This paper will discuss the preparation of can standards. The can standards consisted of four sets; each set comprised four standards with masses ranging from 10 g to 250 g of plutonium.

CAN STANDARDS PREPARATION PROCEDURE

Container

Because the typical collimator height in the can SGS is 12.7 mm, the ideal inner can would be approximately 100 mm in diameter and 250 mm high, with the outer can slightly larger. After an extended search, we concluded that these cans have to be custom designed and built. The fabrication included designing a mold for stamping the lids for the outer and inner cans. After the lids were made, the sides of the can were cut from stainless steel sheets and laser welded to form a can. The sides of all the cans were cut from 0.4-mm-thick stainless steel (SS) and the lids were cut from 0.6-mm-thick SS. The height of the inner can was 273 mm, the diameter was 95.4 mm, and the outer can was slightly larger. The standards are designed so that they can fit into a slip-top can for glove box usage. Figure 1 shows the cans for the standards before the lids are welded.

Matrix

Both graphite and diatomaceous earth have been used in the past as diluent to make SGS standards. Graphite is flammable; diatomaceous earth is used routinely in the glove box as a filtering agent. The density of the diatomaceous earth is also lower (0.26) than graphite (1.25). We selected the diatomaceous earth matrix. The particle size of the diatomaceous earth is measured by sieving to be between 75 μm to 106 μm.

Plutonium Oxide Preparation

During the preparation, the plutonium oxide (containing ~3 kg of plutonium) had been high-fired (900°C), blended for more than 4 hours, and sieved through a 100 mesh sieve (<150 μm). Samples of the oxide standards were analyzed by three different laboratories, New Brunswick Laboratory, Mound Laboratories, and Los Alamos National Laboratory.

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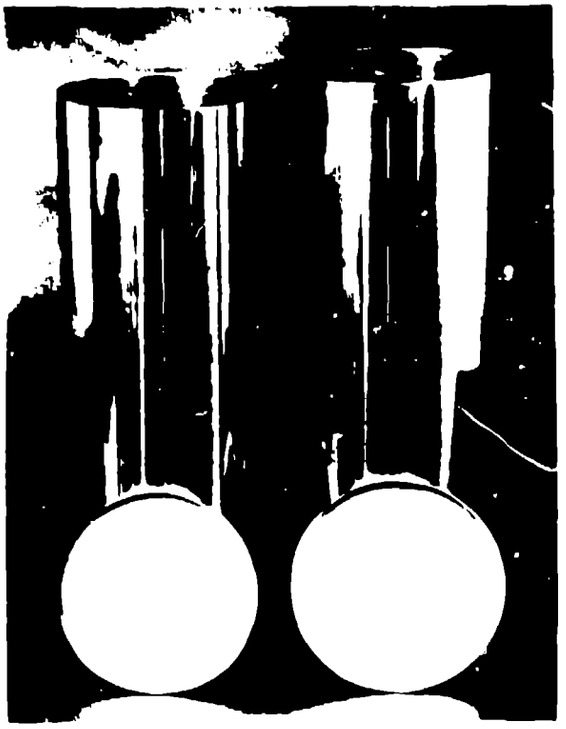

Fig. 1. Can used for the SGS standards before the fabrication.

Details of the chemical analysis will be shown later in the characterization section.

Chemical Characterization

From the blended plutonium oxide batch, two samples were obtained from different parts of the batch for chemical analysis at each of the three laboratories. The analysis determined the plutonium isotopic composition, the grams of plutonium per grams of sample analysis, and the loss on ignition determination. The purpose of the multilaboratory analysis was to minimize the bias from the chemical assay from any of the laboratories; the number of samples would also test the homogeneity of the batch. The results from the Mound Laboratories have been previously reported.[3] Table I shows the plutonium concentration results from the three laboratories. All analysis systems in the three laboratories were calibrated with National Bureau of Standards (NBS) materials and/or certified reference materials.

We observed that the plutonium concentration measurements from the three laboratories agreed closely. This implied that the batch was uniform and that the assay results from the individual samples can be applied to the whole batch. The above chemical analyses were performed in 1986 whereas the actual preparation of the SGS standards took place in 1989. To assure that the plutonium oxide had not changed in this time, another three samples were analyzed by LANL for isotopic distribution and plutonium concentration.

Table I. Chemical Analysis of Plutonium Concentration from the Three Laboratories

LANL		MOUND		NBL	
Sample ID	g Pu/Sample (%)	Sample ID	g Pu/Sample (%)	Sample ID	g Pu/Sample (%)
3-A	87.704	2-A	87.804	8-A	87.722
	87.760		87.816		87.727
3-B	87.745	2-B	87.806	8-B	87.559
	87.673		87.827		87.619
6-A	87.784	7-A	87.818	17-A	87.874
	87.760		87.796		87.798
6-B	87.857	7-B	87.890		87.826
	87.815		87.857	17-B	87.771
					87.800
Average	87.762	Average	87.827	Average	87.737
St dev	0.058	St dev	0.032	St Dev	0.105
RSD(%)	0.067	RSD(%)	0.036	RSD (%)	0.120

Comparison:		
	MOUND/LANL	1.00073
	NBL/LANL	0.99971
	NBL/MOUND	0.99898

All samples decay corrected to 7-1-86

The results were in excellent agreement with the previous analyses. Earlier, more extensive, analyses (1986) were used to calculate the plutonium concentration.

All three laboratories also determined the isotopic distribution with a mass spectrometer. The plutonium isotopes in weight percent (average from the three laboratories) as of July 1, 1986, were

^{238}Pu	0.006
^{239}Pu	96.302
^{240}Pu	3.562
^{241}Pu	0.111
^{242}Pu	0.018

For the SGS standard, the most important isotope is ^{239}Pu. For this isotope, the comparisons among the three laboratories on the ^{239}Pu weight percent were

MOUND/LANL	0.99995
NBL/LANL	0.99998
NBL/MOUND	1.00003

Standard Preparation

Each standard was prepared by putting the plutonium oxide into the inner can, adding the diatomaceous earth, adding helium gas, and welding the lid. After the second outer container was sealed, the cans were helium tested for leaks. Next, the standards were mixed in a V-blender. The can was 60% filled with diatomaceous earth. A blending time of 6 to 8 hours was necessary to assure sufficient mixing.

VERIFICATION

After these standards were prepared, they were measured with an SGS to determine the vertical uniformity. This was performed by determining the quantity of each segment from

the bottom of the can to the top using the 414-keV gamma peak. Attenuation correction was performed for each segment by means of the transmission measurement through that segment. The count time for each segment was 40 s, the collimator was 1.27 cm, and the step size was 0.635 cm. A typical vertical scan is shown in Figs. 2 and 3. From the vertical scans, we found that three to four of the standards were not sufficiently mixed. These were remixed and scanned again. From these vertical scans, we concluded that all the standards are reasonably uniform up to 13 cm, and there are no signs of plutonium and diatomaceous earth agglomerating into clumps observable with gamma scanning.

The same SGS uniformity measurement could also determine ^{239}Pu assay values provided the system was calibrated with known standards. We have two standards, STDASH-1 and STDASH-2, which were prepared 15 years ago, which have been studied extensively in several round robin exercises. These two standards were recently recertified by means of calorimetry and gamma isotopic measurements. Figure 4 shows the comparison between chemical known values and the SGS assay values.

We found that the standard deviation of the ratio of SGS/chemistry was 1.1% with an average of 0.997. This showed that there is no apparent bias between these new standards and the STDASH-1 and STDASH-2. It should be pointed out that these verification measurements were carried out over a six-month period; whenever several standards were prepared, the SGS system was calibrated and the can standards assayed. Subsequent, more careful, measurement showed significant improvements; Fig. 5 below shows the calibration data with one set of these standards for which the standard deviation was ~0.13%.

PARTICLE SIZE

During the preparation, we performed an experiment to determine the particle size of the plutonium oxide. We found that all the plutonium oxide passed through the 100 mesh sieve (150 μm), but only 4% of the oxide passed through the 120 mesh sieve (125 μm).

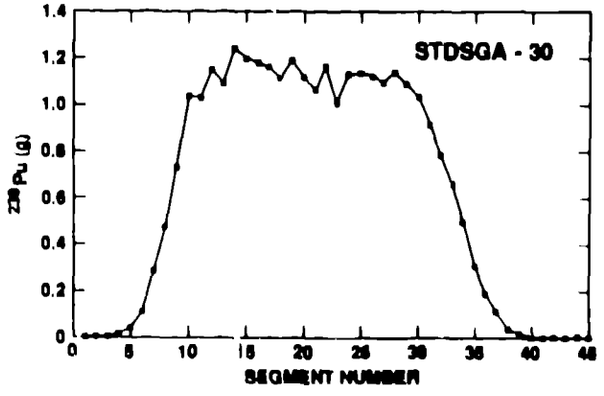

Fig. 2. Vertical scan of the standard STDSGA-30. Each vertical segment increment is 0.635 cm.

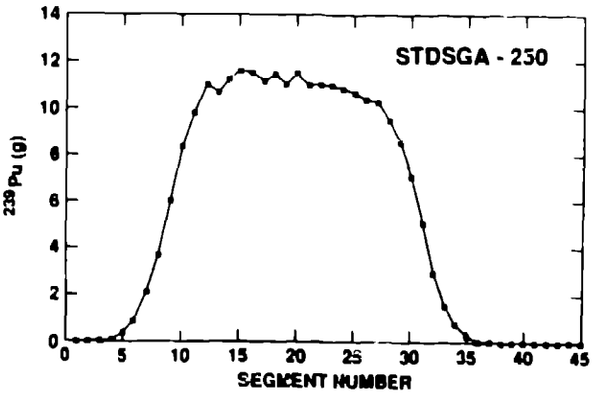

Fig. 3. Vertical scan of the standard STDSGA-250.

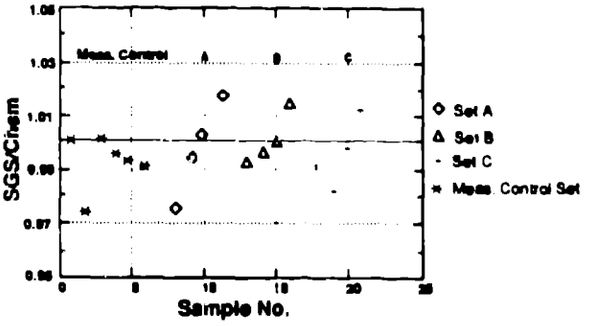

Fig. 4. Verification measurements by the SGS technique for all the standards as compared with chemical preparation values.

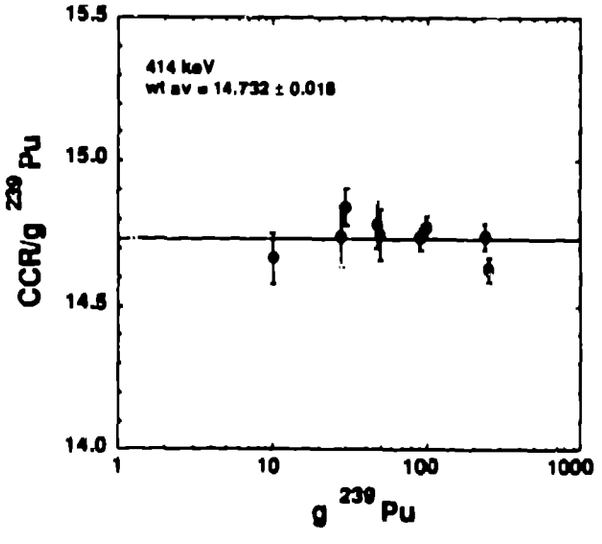

Fig. 5. Measurements for set A of the standards plus one of the measurement control standards (STDSGMC-6).

During the Fast Flux Test Facility oxide program in early 1980, when we had the particle size analyzer at Los Alamos, we found that the mean oxide particle was 5 μm in size, but could only pass through a 100 to 120 mesh sieve. We believe that the oxide particles are small but they tend to agglomerate into large clumps; the suspension technique of particle size determination tends to break down the agglomeration. At worst, the plutonium particles have a diameter of 150 μm and a density of 10 g/cm^3; at best the particle size is ~5 μm. The most likely particle size is somewhere in between. Table II shows the leakage fraction from plutonium oxide particles of different sizes at 413 keV, the usual assay energy for plutonium, assuming the oxide density of 10 g/cm^3.

Table III shows the final plutonium content of these standards as of January 1, 1990.

CONCLUSION

The paper shows that careful consideration needs to be given to the preparation and quality control of well characterized SGS standards. Documentation is extremely important; without it the standards are incomplete. The procedures described in this paper were used to successfully prepare 4 sets or 20 SGS standards, which meet the physics requirements of the measurement as well as the regulatory requirements of the new DOE order.[4] They are being used daily at Los Alamos and the Westinghouse Savannah River Site.

Table II. Gamma-Ray Leakage Fractions from Plutonium Oxide Particles of Different Sizes

Particle Size(μm)	Mesh Size	Leakage at 413 keV
150	100	0.985
106	140	0.989
90	170	0.991
75	200	0.992
53	270	0.994
38	400	0.996
20		0.998
5		1.000

Table III. The Final Plutonium Content of these Standards as of January 1, 1990

Standard ID	^{239}Pu (g)	Uncertainty (g)	(%)
STDSGMC-1	48.142	0.035	0.073
STDSGMC-2	48.130	0.035	0.073
STDSGMC-3	48.131	0.035	0.073
STDSGMC-4	48.132	0.035	0.073
STDSGMC-5	48.139	0.035	0.073
STDSGMC-6	48.130	0.035	0.073
STDSGA10	9.629	0.012	0.125
STDSGA30	28.874	0.023	0.078
STDSGA100	96.269	0.068	0.071
STDSGA250	240.661	0.169	0.070
STDSGB10	9.624	0.012	0.125
STDSGB30	28.890	0.023	0.078
STDSGB100	96.273	0.068	0.071
STDSGB200	192.528	0.135	0.070
STDSGC10	9.625	0.012	0.125
STDSGC30	28.876	0.023	0.078
STDSGC100	96.268	0.068	0.071
STDSGC250	240.664	0.169	0.070
STDSGCAL20	19.271	0.017	0.087
STDSGCAL200	192.527	0.135	0.070

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4. Department of Energy Order 5633.3.

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