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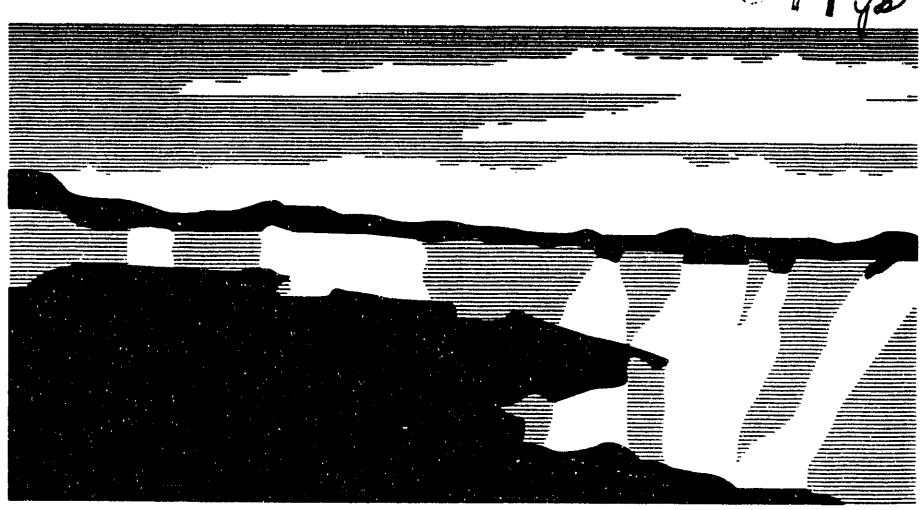

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DESIGN AND FABRICATION OF THE URANIUM DRUM STANDARDS*

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ABSTRACT

The segmented gamma scanner has become an important instrument for assaying the special nuclear material content of low-density scrap and waste in 55-gal. drums. To perform these assays accurately, we need appropriate standards to calibrate the response of the system including the detector efficiency, the absorber in front of the detector, and the collimator geometry. A set of three uranium drum standards has been fabricated at Los Alamos. This paper discusses a Monte Carlo calculation to optimize the design of the drum standards. The drum standards are prepared using 20 modular 4-ℓ bottles in each drum. This paper also describes the fabrication procedure, which includes weighing the uranium oxide and the chemical analyzing of the uranium concentration. Also presented is the vertical scanning data of the 4-ℓ bottles to assure uniform mixing of the uranium and the diluent. Finally, the nondestructive measurements for checking consistency among the three drum standards are discussed.

INTRODUCTION

Los Alamos National Laboratory (LANL) is using several segmented gamma scanners (SGS) to assay the special nuclear material (SNM) in scrap and waste in 55-gal. drums. To perform these assays accurately, one must have appropriate standards covering the range of the samples to calibrate the systems. The need for a set of uranium drum standards to calibrate the SGS instruments was identified earlier by the DOE auditors and in 1990 by the Standard Advisory Committee at LANL. The work was initiated in late 1991 and was financed by the Safeguards and Security Upgrade funds. It was a team effort with members from four different groups contributing to the design, fabrication, and verification of this set of standards. The drum standards were prepared by September 1992; they were verified more recently.

We have followed the American National Standards Institute (ANSI) guidelines for the general preparation of SGS standards.[1] It is not necessary for the standards to duplicate the chemical form or composition of the unknown samples; however, the standards should be designed to meet the following requirements.

1. The SNM should be reasonably uniformly distributed in the standards.

2. The standard should have a diameter so that the gamma-ray transmission through the standard is reasonable ($0.2 < T < 0.6$).

3. The height of the standard containing SNM should be at least several times the height of the collimator used in the SGS measurement to minimize the end effect.

4. The particle size of the SNM should be small so that the self-absorption of the particle is negligible at energies as low as 203 keV.

DRUM STANDARDS DESIGN

Approach

Because of the size of the 55-gal. drum, it is difficult, if not impossible, to mix the SNM directly in the drum to achieve uniform distribution. The more practical approach is to mix the SNM with the low-Z matrix in 4-ℓ bottles and distribute the bottles in the drum so that they best represent a uniform distribution. However, there are questions as to how to position the bottles in the drum and whether the voids between the bottles should be left empty or filled with the matrix.

Monte Carlo Study

It was decided to use the Los Alamos Monte Carlo transport code to study the best configuration for the SGS drum standards. Three cases were examined: (a) SNM uniformly distributed in the drum as the reference, (b) SNM contained in seven columns with void between the columns, and (c) SNM contained in seven columns with diatomaceous earth filling the void between the columns. In all cases, 105 g of plutonium is contained in the drum and the gamma-ray energy is 414 keV. Table I gives the

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TABLE I. SGS Barrel Standard Calculation Study Results

	Normalized Count Rate	Transmission	Correction Factor [a]	Total Corrected-Count Rate
a. Uniform	1.00	0.1975	1.769	1.00
b. Columns +Void	1.25	0.3448	1.476	1.04
c. Columns $+SiO_2$	1.01	0.1973	1.769	1.01

[a]Correction factor defined as $CF = (-\ln T^{p/4})/1 - T^{p/4}$.

results of the study.[2] In case (b), the total corrected count rate is 4% higher than the reference case. This is mainly because the normalized count rate has increased by as much as 25% compared to the uniform case. The situation that most closely resembles the uniform case is when the gaps between the columns of bottles containing the SNM are filled with the matrix material, case (c); the total corrected count rate is within 1% of that of the uniform drum. Similar conclusions can be reached for gamma-ray energy of 186 keV.

Final Configuration

It is the conclusion of this study that the drum standards should be prepared with twenty 4-ℓ bottles to best simulate the columns and that the gaps between the bottles should be filled with Cellutex. The configuration of the bottles in the drum is shown in Fig. 1 (side view) and Fig. 2 (top view). Cellutex was selected to fill the gaps between the bottles because the density of Cellutex (0.26 g/cm^3) is almost identical to that of diatomaceous earth and has similar transmission properties. Because of the rigidity of Cellutex, it can be used to hold the bottles securely in place and yet allow the possibility of removing the bottles for future mixing or other studies.

FABRICATION OF THE DRUM STANDARDS

It was decided to fabricate three drums containing nominally 30, 100, and 200 g of ^{235}U. A procedure to fabricate the drum standards was first developed by the team members, and the final version was approved by the Standard Advisory Committee in February 1992. The document outlined the procedural steps and was followed closely by the members who prepared the standards. The main stages in the preparation process are described below.

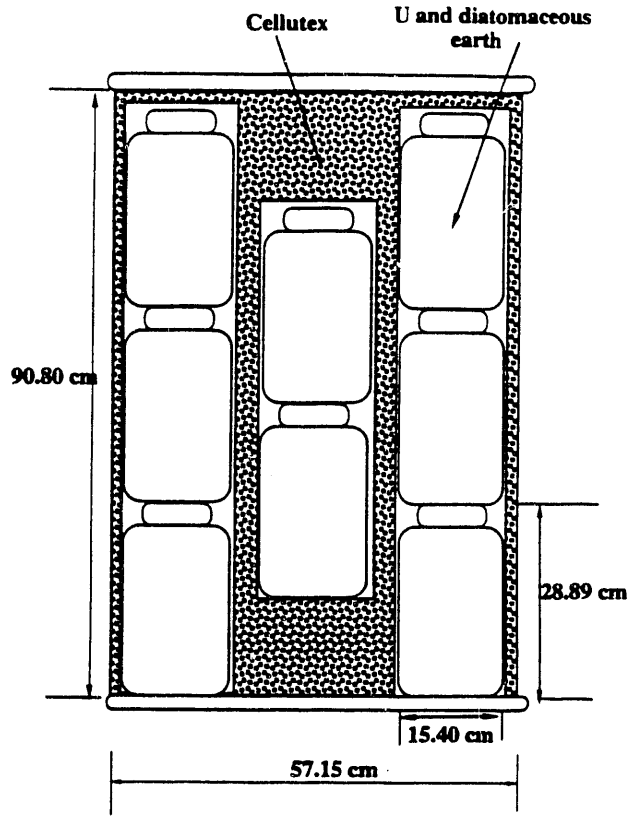

Fig. 1. Side view of the 4-ℓ bottles in the drum in Cellutex matrix.

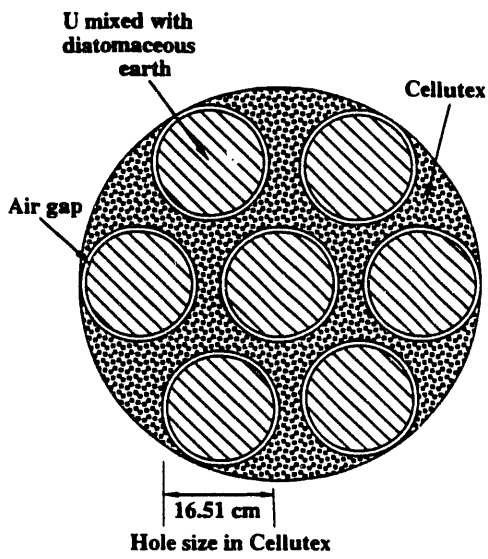

Fig. 2. Top view of the 4-ℓ bottles in the drum.

Preparation of Drum

Normal 55-gal. drums are too short to accommodate 4-ℓ bottles stacked three high. Each drum was lengthened 6.35 cm by welding two drums together. Leak tests were performed on all three drums. The Cellutex, each piece precut with seven 16.51-cm-diam holes, was stacked in the drum and empty bottles were positioned in the drum to test for fit. Three drums and 60 pieces of Cellutex were prepared according to the specification in Figs. 1 and 2.

Preparation of Material and Chemical Characterization

The standards were prepared from relatively pure uranium oxide (U_3O_8) diluted with diatomaceous earth. Approximately 1.5 kg of the feed oxide was high-fired to 900°C in a furnace for 2 h. It was sieved through a 100-mesh sieve to produce a particle size less than 150 μm and blended in a Turbula blender for approximately 1 h. Five samples taken from different parts of the blended batch were analyzed by the Analytical Chemistry Group at LANL. The chemical analysis determined the uranium isotopic composition, the uranium weight fraction, and the loss on ignition.

The analysis systems used for chemical determination were calibrated with National Bureau of Standards materials or certified reference materials, or both. Results of the chemical assay for the five samples are summarized in Table II. As shown in the table, the uranium concentration measurements agreed well with the quoted accuracy of the analytical method, and the isotopic distributions for ^{235}U were in excellent agreement among the five samples. This implies that the batch was uniform and that the assay results from the individual samples can be applied to the whole batch.

TABLE II. Summary of Analytical Data for Uranium Oxide

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
Av	84.33	92.18	92.11	
St. Dev.	0.039	0.009	0.005	
RSD (%)	0.05	0.01	0.01	

Weighing, Blending, and Loading

A LANL-certified balance with 3-digit readout was used throughout the weighing of the feed material and associated apparatus. All weighings were recorded on data sheets and verified by a second person as specified in the procedure.

The prelabeled 4-ℓ bottle and the plastic weighing boat were tare weighed separately. A required amount of oxide was weighed in the plastic boat and transferred into the bottle. The holdup in the boat was determined by reweighing after the contents were removed. The bottle with the contents was weighed to verify the amount of U_3O_8 introduced. Diatomaceous earth was added to the bottle until it was approximately 80% full and then it was weighed again. The bottle with a lid taped on was labeled and its gross weight determined. These steps were repeated for the remaining 19 bottles for the drum.

Each bottle filled with the mixture was blended in the Turbula blender for 60 min and measured with the SGS to test its uniformity. Results of these tests are discussed in the next section. After 20 bottles had passed the uniformity test, they were loaded into the drum as shown in Fig. 1. The locations of the bottles in the drum were marked and a LANL label and TID were applied.

By following this preparation procedure, we fabricated three uranium drum standards in the ranges of 30, 100, and 200 g of ^{235}U. See Table III for the final uranium contents of these standards and the estimated uncertainties.

VERIFICATION

After all the 4-ℓ bottles were prepared, they were scanned, using the SGS, for vertical uniformity determination. This was performed by measuring the total corrected counts (TCC) in each segment, from the top of the bottle to the bottom, using the 186-keV gamma peak. The attenuation of each segment was corrected by calculating the correction factor from the transmission. A typical vertical scan of the bottle is shown in Fig. 3. From the vertical scans, we found that eight of the standards were not sufficiently mixed; they were reblended and rescanned.

TABLE III. Consistency of the Three Drum Standards			
Run	STDUD1-30 g TCC	STDUD2-100 g TCC	STDUD3-200 g TCC
1	1088.48	3528.55	6979.60
2	1094.80	3531.86	6965.72
3	1081.20	3533.20	6929.62
4	1099.10	3548.61	6949.59
5	1083.81	3543.65	6970.58
6	1089.53	3549.84	6924.21
7	1085.91	3528.35	6933.53
8	1086.22	3548.82	6935.04
9	1096.23	3537.07	6969.68
10	1092.86	3541.42	6971.26
Av TCC	1089.81	3539.14	6952.88
Known ^{235}U (g)	31.198	100.987	200.428
Uncertainty (g)	0.058	0.187	0.372
Av TCC/^{235}U	34.932	35.045	34.690

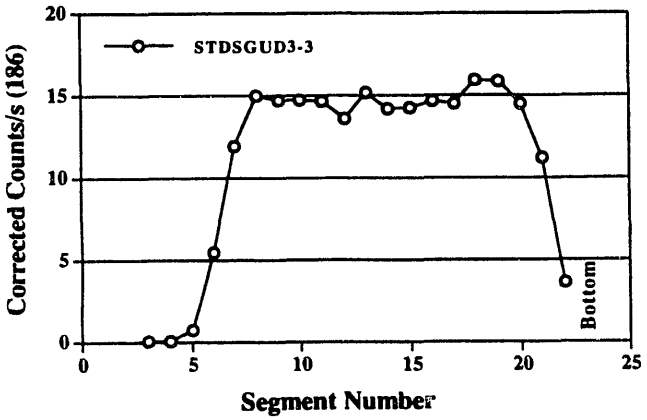

Fig. 3. Profile of the 4-ℓ bottle.

After the standards were labeled and loaded, an SGS drum assay system was used to study the consistency response among the three drum standards. Each of the standards was measured 10 times using the SGS system. The averaged counts, corrected for rate loss and attenuation at 186 keV, were calculated and tabulated in Table III. The averaged corrected counts divided by the known ^{235}U contents for the standards are also listed in Table III. These ratios, representing the calibration constants for the SGS system, are plotted as a function of the standards in Fig. 4. As shown, the three calibration constants agree to 1%. This indicates that there is a relatively high degree of consistency among the three standards, and any one of the three can be used to calibrate the SGS systems to 1%.

The 30- and 100-g standards were also used as control checks of the SGS system. These standards were measured daily in alternate mornings and afternoons over a period of six weeks. Figure 5 shows the ratios of assayed to known values as a function of dates measured for both standards. The reproducibility of both standards is 0.53%. This is in agreement with the counting statistics of 0.67% and 0.43% for the two standards, respectively. The data demonstrate that both the standards and the SGS measurement system are stable.

CONCLUSION

We have successfully fabricated a set of three uranium drum standards using the above procedures. These standards meet the physics requirements of the measurements as well as the regulatory requirements of the DOE Order.[4] From our experience, we have learned that standard preparation is very costly and time consuming. It requires thorough planning, attention to details, coordination, and proper supervision in all stages of preparation. Finally, documentation is extremely important; without it the standards are incomplete.

ACKNOWLEDGMENTS

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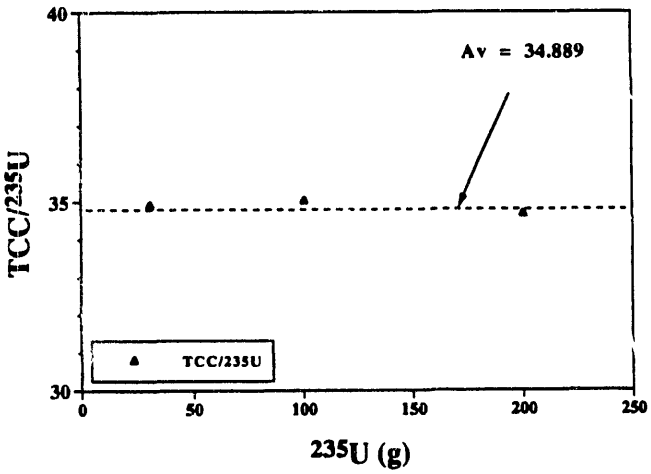

Fig. 4. Consistency of the three drum standards. The calibration constants from the three standards agree to 1%.

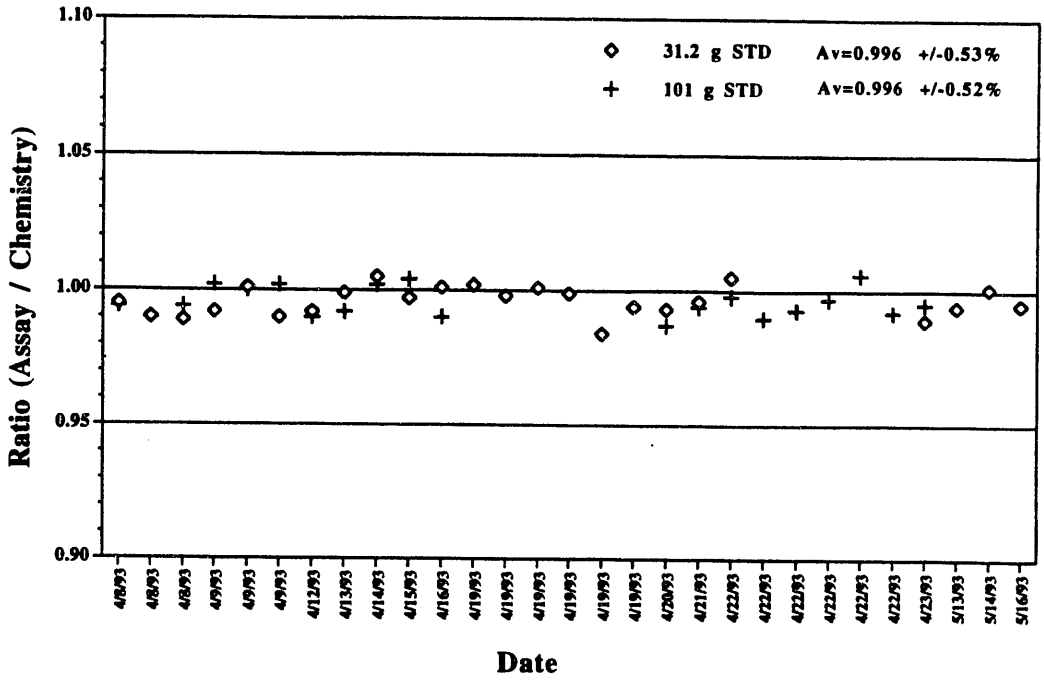

Fig. 5. Reproducibility of the standards over a period of time.

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