

Measurement of PDP Drums Evaluate Program Isotopic

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Introduction

Measurement of nonhomogeneous special nuclear material using passive gamma techniques is difficult because it is not practical to develop standards with the characteristics of the samples. Each sample is different. And the samples must be measured nondestructively. Modeling methods must be used. The modeling methods assume homogenous distribution of radioactive and matrix material within the container. In practice, the activity must penetrate localization matrix attenuation or attenuation by the heavy material itself. In addition, geometry errors are introduced when the activity is localized in one spot.

Program description

A Performance Demonstration Program¹ (PDP) was established in 1998 for transuranic (TRU) waste characterization for the Waste Isolation Pilot Plant. Under this plan, blind audit samples were used as an independent means to assess the performance of nondestructive assay (NDA) measurement groups regarding compliance with established Quality Assurance Objectives. Samples and drums used for this program consisted of 208-liter (55-gallon) drums with fabricated matrix inserts and a defined set of standards of uranium and plutonium. The sources of plutonium were composed of 93.8% ²³⁹Pu and uranium sources were enriched to 93% ²³⁵U. The uranium and plutonium was absorbed in diatomaceous earth to minimize gamma-ray self absorption from these heavy elements and encapsulated in cylinders 9 inches tall and 1.7 inches in diameter. The drums contained tubes that allowed the cylinders of samples can be positioned at different vertical locations. As part of the PDP program, rods were prepared to simulate matrices of glass, mixed metals, and combustible material. The empty drums contained brackets to hold these rods.

In May 2002, at Los Alamos National Laboratory a program was developed and implemented by Mike Baker of Los Alamos National Laboratory to assist ORTEC in the evaluation of its portable nondestructive assay system. The hardware portion of the assay system chosen for this evaluation consisted of the ISO-CART^a and the software programs consisted of GammaVision and ISOTOPIC^b.

Drums and material from this program were used to provide a wide variety of sample simulations. Samples of plutonium varying from 0.027 to 50.31 g of ²³⁹Pu were selected. The simulated drums contained from one to four sources distributed at random positions throughout the drum. The total ²³⁹Pu sample loading varied from 0.056 g to 89.1 g. The total ²³⁵U loading varied from 4.987 g to 24.97 g. The sample position and loading values were kept from the analyst until the measurements and analyses were completed. Four matrix drums were prepared that contained rods representing glass, combustible, metals, and noninterfering (blank) materials. The density of each of these drums did not change within the material type throughout the measurement exercise.

Description of data acquisition and analysis software

The GammaVision software package was used as the basic gamma-ray spectroscopy measurement tool. This software was used to develop a gamma-ray library, establish detector efficiency, acquire the data, determine accurate peak areas, and produce an extensive report after the acquisition was complete.

^a Portable high-purity germanium gamma-ray spectroscopy system marketed by ORTEC

^b Radioactive waste characterization software licensed from Lockheed Martin and distributed by ORTEC

Modeling software was used to simulate the waste drums. The software used for this step was called ISOTOPIC. ISOTOPIC² was designed to use the activity computed from the GammaVision report and correct for geometry, matrix, container, plutonium, uranium, and air attenuation and then print an extensive report of the results. These correction factors were applied to all gamma rays detected in the spectrum, however, only one reference gamma rays was used to report the results. Fine-tune adjustments to the matrix density or container thickness can be made after the initial correction factors are applied. The activity results were converted to grams within this program. A more extensive description of the correction factors is given in Reference 2.

System calibration

The data acquisition system was calibrated using a point source traceable to the National Institute of Standards and Testing. This standard contained a wide energy range of gamma rays whose emission rates were well established. The point source was positioned 30 cm from the face of the detector and counted for several hours to obtain excellent counting statistics. The efficiency response established for the detector was then implemented within the GammaVision software. Thus, GammaVision reported activity as if the items that it measured were point sources. These point-source activities were then corrected for the characteristics of the item being measured using Program Isotopic.

Gamma-ray library development

Libraries containing information about expected nuclides were developed using the gamma-ray branching ratios for every gamma ray expected in the spectrum. Reference gamma rays were selected for each nuclide. The activities of all the gamma rays are determined, but only the reference gamma ray was used for the reported results. The best reference gamma rays selected for reporting were gamma rays that are high energy, intense, and free from interference. In practice, many high-energy gamma rays are not intense and compromises are made between high energy and intensity. The reference gamma ray used for ²³⁹Pu analysis was the 413.7-keV gamma ray. This is not the most intense gamma ray emitted during ²³⁹Pu decay, but its medium energy is more penetrating than the more intense 129.3-keV gamma ray. The reference gamma ray at 185.7 keV was used for the ²³⁵U quantification.



Fig. 1 ISO-CART measurement system

Description of the NDA hardware

A portable HPGe detectors with an efficiency of 80 % efficiency relative to an 7.6-cm × 7.6-cm (3-in. × 3-in.) NaI detector, was used to collect the data. The detector was positioned be at the center of the barrel or box using an ISO-CART. A picture of an ISO-CART is shown in Figure 1. The multichannel analyzer used to collect the data is called a digiDart. This digiDart is pictured with the ISO-CART in Figure 1.

Measurement description

The drums were moved into the measurement position with a fork lift as shown in Figure 2. They were then scanned for localized activity as shown in Figure 3. Counting times varied from 15 minutes to 1 hour depending on the concentration of activity and time available. The detector was positioned approximately 18 inches away. The vertical position of the detector was halfway from the bottom of the container. The items were rotated on a turntable at a speed of 1 revolution /minute.



Fig. 2 Drums were positioned with a fork lift



Fig. 3 Drums were scanned for localized activity

The four drums containing rods simulating the four matrices were loaded with different quantities of sources. Sixteen items were counted with different ranges of activity.

Results

The results of the containers were presented to the program director. The measured results, R_{meas} , were then compared with the prepared results, R_{prep} . The comparison percentage, R_{com} , was then computed using the following equation:

$$R_{\text{com}} = (R_{\text{meas}} - R_{\text{prep}}) * 100 / R_{\text{prep}}$$

The results are shown in Table 1.

Discussion of Results

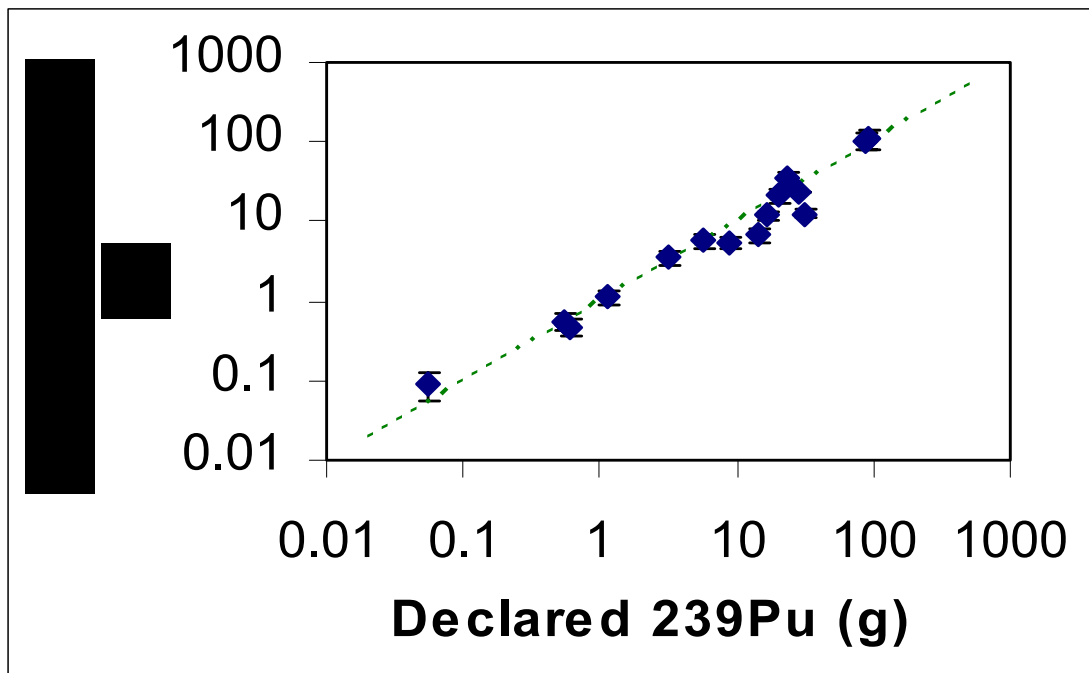
For most of the samples of ^{239}Pu agreement was excellent from the low loadings to the highest loadings as indicated in the plot. Except for two ^{239}Pu outliers and one ^{235}U outlier, the average differences are within 21 per cent. This is the average estimated uncertainty reported for the plutonium results. Thus, the reported uncertainty levels are very realistic.

Results of the outlier measurements were reviewed. The low results of drum #13 can be explained, because one of the samples was positioned on the outside of the container. This was caught by the prescreening. Thus, the sample was not rotated, and the sample was analyzed assuming all the activity was in that sample. Unfortunately, another sample was positioned in the middle of the drums. This sample was not detected but it contributed to the overall activity. The geometry correction factor should have been much greater.

The other two outliers, Drums #3 and #14, were rotated as because no definitive localized activity was detected. The analysis results were thoroughly reviewed and the discrepancies could not be explained.

Table 1. Measurement Results of PDP Drums

Drum #	Matrix	Declared ²³⁹ Pu (g)	Declared ²³⁵ U (g)	Measured ²³⁹ Pu (g)	Est'd Uncert.	Compare ²³⁹ Pu	Assay ²³⁵ U (g)	Est'd Uncert.	Compare ²³⁵ U
1	COMBUST	0.574		0.502	+/-16%	-13%			
2	NONINT	8.970		5.57	+/-16%	-38%			
3	METALS	14.110		6.88	+/-19%	-51%			
4	GLASS	1.110		1.12	+/-22%	+1%			
5	METALS	5.604		5.79	+/-19%	+3%			
6	COMBUST	89.121		103	+/-22%	+16%			
7	GLASS	16.851	4.987	11.8	+/-10%	-30%	4.94	+/-10%	+1%
8	COMBUST	3.151		3.46	+/-18%	+10%			
9	NONINT	23.618		34.5	+/-20%	+46%			
10	GLASS	0.604		0.48	+/-22%	-20%			
11	COMBUST	19.715		21.1	+/-22%	+7%			
12	METALS	0.536		0.564	+/-22%	+5%			
13	COMBUST	30.770		12.6	+/-10%	-59%			
14	GLASS	28.451	24.976	24.1	+/-27%	-15%	67.4	+/-38%	+170%
15	GLASS	0.056		0.089	+/-38%	+58%			
16	METALS	93.367		108.7	+/-29%	+16%			
Ave					21%	24%			



Lessons learned

It is assumed that the outliers represented items that were positioned close to the outside of the container. This error can be reduced by positioning the detector further from the item being measured when plenty of activity is present. Thus, a standard standoff position of 24 inches will be utilized. In the future if more sensitive TRU measurements are required, this standoff can be reduced to 18 inches.

Acknowledgement

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References

¹ DOE, Performance Demonstration Program Plan for Nondestructive Assay for the TRU Waste Characterization Program,” CAO-94-1045, Revision 1, Carlsbad Area Office, Carlsbad, NM, May 1998.

² Hagenauer, R.C., “A Portable HPGe System for Measuring Contaminated Soils and Floors,” Proceedings of the International Conference on Nuclear and Hazardous Waste Management Spectrum 2000, Chattanooga, TN Sept 2000.