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TITLE NONDESTRUCTIVE ASSAY OF PLUTONIUM BEARING SCRAP AND WASTE WITH THE ADVANCED SEGMENTED GAMMA-RAY SCANNER

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ABSTRACT

Assaying plutonium-bearing scrap and waste (S&W) for plutonium content can be very difficult because of the heterogeneous nature of the items. Previous efforts have been hampered by the lack of representative standards for calibrating and evaluating measurement performance on actual plant materials. We have characterized 25 S&W items in three distinct S&W categories to 2% or better. We used these items with fabricated calibration standards to evaluate the performance of the lump-corrected segmented gamma-ray scanner. We show that some difficult-to-measure S&W samples can be assayed with less than 10% bias, but still suggest that each category of S&W be individually evaluated for measurement bias.

INTRODUCTION

All nuclear material processing facilities generate large amounts of heterogeneous scrap and waste (S&W) containing special nuclear material (SNM). This scrap and waste needs to be measured to compute the material balance. (We define scrap as material with sufficient SNM to make recovery economically viable; waste does not have sufficient SNM for recovery.)

Most chemical techniques rely on sampling the item and then extrapolating the results from the sample to the original item. For heterogeneous scrap and waste, chemical analysis is not viable because of the difficulties of obtaining representative samples; these samples have traditionally been measured by nondestructive assay (NDA) instruments. NDA has some advantages compared to chemical analysis; it measures the entire sample, the sample does not have to be homogenized, it does not generate more waste, and it is easier to apply. However, NDA is not the answer to all assay problems; some waste, for example, large crates, cannot be measured quantitatively by any means.

This paper describes the capability of the advanced segmented gamma-ray scanner (SGS) to measure a wide variety of plutonium-bearing scrap and waste. Real samples were obtained from operating facilities and subsequently carefully characterized. The samples include low-density plutonium bearing ash, high-density plutonium oxide, sand-slag crucibles (SSC), and salts generated from the molten salt extraction (MSE) process. This paper demonstrates that some of these process samples can be measured quite well with the state-of-the-art techniques on the SGS.

PRINCIPLE

The SGS[1] was developed in the 1970s for low-density, reasonably uniform samples. The SGS assay is based on the 413-keV gamma ray spontaneously emitted from ^{239}Pu. Corrections are made for rate-related counting losses and the attenuation from sample matrix self-absorption. The sample is viewed in slices or segments defined by moving the sample vertically, in discrete steps, in front of a collimated detector. The segmentation reduces the bias caused by vertical heterogeneity in the sample. The samples are rotated during the assay, which tends to reduce the effects of horizontal heterogeneity.

Assay results, however, can still be biased (nearly always low) if the density of the sample is high or if the sample contains lumps of plutonium. Recent improvements in SGS analysis started with improving the matrix attenuation correction factor, and evolved into correcting for the presence of the lumps.[2] These improvements can be summarized as follows:

1. Quadratic interpolation of the measured transmission to deduce the sample transmission at the assay energy rather than use the transmission measured at the transmission source energy.[1]

2. Calculation of the attenuation correction factors using numerical integration of actual sample/standard geometry instead of the far field approximation. Polynomial functions are used to fit the correction factors as a function of the transmissions. This improvement reduces some of the correction factor errors for homogeneous samples from between 5% and 10% to 1%, using numerical integration as the reference and benchmarking with measurements on standards.

3. Assays for plutonium are performed at several energies, and the suppression of the lower energy assay is used as an indicator of the presence of lumps of SNM in the sample. A lump correction is performed based on the assumption that the lumps are spherical and of the same size.[2]

These improvements in the data analysis are the main features of the advanced SGS. To test its validity, the instrument was used to assay a group of well-characterized process samples whose SNM contents are characterized to 2% or better by methods described below. The results of the advanced SGS assays are compared with a neutron coincidence counter (NCC). Traditionally, for the plutonium bearing S&W, we

suspect the results from the NCC are biased high, and that the SGS results are biased low. This can be used, in most cases, to bracket the true result.

CALIBRATION

The advanced SGS was calibrated with the standard STDASH-1; the calibration was validated with STDASH-2. Both of the STDASH standards are fabricated from plutonium oxide and diluted with diatomaceous earth. We have found these standards to be uniform and homogeneous. The particle size distribution of each standard is not known.

The NCC was calibrated with the PEO and LAO series of oxides. The PEO samples are prepared with 10% ^{240}Pu ranging from 20 g to 780 g of plutonium; the LAO samples are prepared with 16% ^{240}Pu and with plutonium masses ranging from 60 g to 870 g. The plutonium contents of these standards were determined from the chemical preparation and recently validated with calorimetry and gamma isotopics. These standards are intended for the NCC; in general they are too dense for the SGS and fairly opaque for gamma-ray transmission at the assay energy. They mimic clean scrap.

RESULTS

The results are summarized in Table I for both instruments.[3]

The first set of samples measured consists of plutonium-bearing ash with a relatively low density (~0.5 g/mL) from Hanford. These samples are appropriate for the SGS because they are low-density waste that is fairly uniform. Figure 1 shows the SGS assay results. The SGS assays have a relative standard deviation (RSD) of 0.7%, which is approximately the precision of the measurements with very little bias. In comparison, the NCC assays of these samples fluctuate substantially more with an RSD of 5%.

The second set of samples consists of the plutonium oxide. Although these samples have a relatively high density and should be measured by NCC, they were measured with SGS for completeness. The results are shown in Fig. 2. In general these samples are too dense for the SGS and fairly opaque for gamma-ray transmission at the assay energies. The NCC assay results have an RSD of 3% while the SGS assay results fluctuate with an RSD of 24%. The average SGS result is only 6% low: probably a fortuitous result of sample geometry.

Table I. Measurement Results from Well-Characterized Samples

Sample ID	Reference (g Pu)	SGS (g Pu)	Sigma (%)	SGS/Reference Ratio	NCC (g Pu)	Sigma (%)	NCC/Reference Ratio	NCC & SGS Average	Av/Reference Ratio
Low-density Ash									
STDASH - 1 *	20.89	20.91	0.45	0.998	20.13	1.60	0.959	20.52	0.977
STDASH - 2 *	20.85	21.08	1.41	1.005	20.17	1.77	0.963	20.62	0.984
ASH 685-003	33.14	32.98	0.84	0.995	32.53	3.05	0.981	32.75	0.988
ASHHVA - 27	51.30	52.03	0.38	1.014	49.22	4.11	0.959	50.62	0.987
ASHHVA - 5	164.95	164.94	0.43	1.000	176.81	5.00	1.071	170.78	1.035
ASHHVA - 6	6.70	6.72	1.81	1.004	6.91	8.11	1.031	6.72	1.004
			Av =	1.002		Av =	0.994	Av =	0.998
			SD =	0.007		SD =	0.047	SD =	0.021
High-density Oxide									
PEO - 381	313.00	312.98	1.19	1.000	332.48	1.39	1.032	322.72	1.016
PEO - 386	467.89	471.15	4.73	0.998	480.48	1.92	1.045	445.80	0.974
PEO - 447	776.46	798.55	2.18	1.030	832.11	1.09	1.072	815.63	1.051
LAO260C10	69.80	44.37	1.46	0.742	68.88	2.34	0.981	61.53	0.862
PEO - 382A & LAO261C10	78.78	78.87	0.96	0.[illegible]1	78.87	0.89	0.981	78.87	0.961
LAO261C10	171.43	89.84	4.[illegible]2	0.[illegible]2	173.72	5.62	1.013	131.83	0.768
LAO262C10	320.98	384.42	4.89	1.136	321.74	1.62	1.002	343.08	1.069
LAO263C10	611.04	729.78	3.82	1.194	604.22	1.43	0.989	667.00	1.092
LAO261C11	874.10	911.85	0.84	1.043	871.61	1.41	0.997	891.68	1.020
			Av =	0.947		Av =	1.010	Av =	0.979
			SD =	0.[illegible]88		SD =	0.034	SD =	0.105
Sand-Slag-Crucibles									
MPX -1826	134.28	102.84	0.31	0.766	178.24	1.22	1.328	140.54	1.047
MPX -1843	216.00	218.58	0.49	1.013	263.08	1.90	1.218	240.98	1.116
MPX - 1907	222.81	165.14	0.24	0.741	284.77	2.44	1.278	224.95	1.010
MPX - 1945	63.70	52.64	0.39	0.830	68.85	8.98	1.081	60.65	0.955
MPX - 1986	124.00	109.90	0.61	0.886	166.72	3.27	1.256	134.81	1.071
MPX - 2190	166.00	166.32	0.23	0.936	172.21	1.60	1.037	163.77	0.987
MPX - 2240	136.00	133.78	0.28	0.984	163.55	1.84	1.129	143.67	1.056
MPX - 2298	294.00	109.99	0.28	0.374	504.98	1.65	1.718	307.48	1.046
			Av =	0.816		Av =	1.266	Av =	1.036
			SD =	0.204		SD =	0.212	SD =	0.051
Molten Salt Extraction									
XBLP - 121	155.37	148.49	0.38	0.956	238.79	9.75	1.537	148.49	0.956
XBLP - 278	90.38	86.98	0.38	0.951	121.83	12.28	1.348	85.89	0.951
XBLP - 301	248.85	231.37	0.22	0.861	284.33	3.80	1.070	250.85	1.016
RFMSE - 1	243.83	231.62	0.60	0.850	474.67	7.10	1.946	231.62	0.950
RFMSE - 2	372.73	354.80	0.71	0.952	790.74	4.72	2.121	345.80	0.952
RFMSE - 4	408.88	397.89	1.31	0.974	887.34	5.90	2.196	397.89	0.974
ARF878685	263.86	256.74	1.05	0.970	463.97	4.24	1.722	256.74	0.970
ARF878642	219.80	202.70	1.31	0.923	387.43	8.46	1.756	202.70	0.923
			Av =	0.888		Av =	1.712	Av =	0.982
			SD =	0.018		SD =	0.388	SD =	0.027
Savannah River In-plant Standards									
4359	381.17	381.20	0.01	1.000	399.40		1.048	389.80	1.025
4360	374.87	381.20	1.77	1.018	349.49		0.933	365.35	0.975
4361	113.42	112.20	1.08	0.989	114.67		1.011	113.44	1.000
4362	418.38	418.44	0.22	0.998	436.90		1.039	427.17	1.019
			Av =	1.001		Av =	1.008	Av =	1.009
			SD =	0.012		SD =	0.053	SD =	0.022

* Samples prepared with plutonium oxide and diatomaceous earth

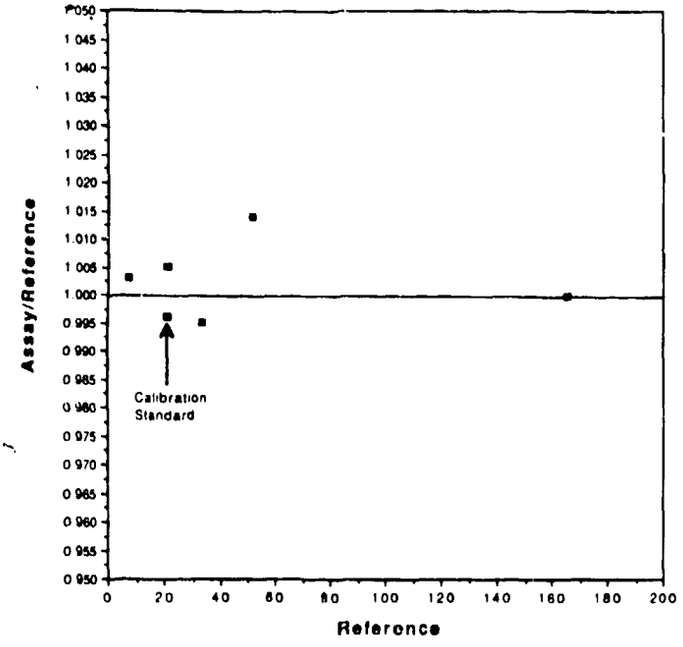

Fig. 1. Assay accuracy of the SGS for ash samples. These samples are uniform and of low density: well suited for SGS assay. The reference values are known to better than 1%.

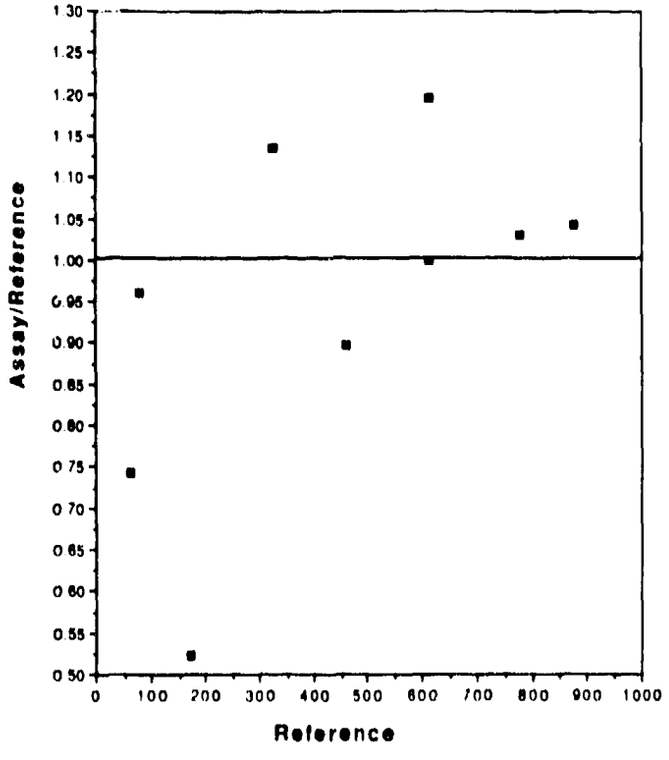

Fig. 2. Assay accuracy of the SGS for the plutonium oxide items. These pure, high density samples have well documented chemical compositions. The reference values are known to a few tenths of a percent.

The third set was the SSC samples generated in the pyrochemical process: residues from plutonium casting and broken crucible pieces. These samples are relatively high-density waste, with a low SNM mass heterogeneous in both the matrix and the plutonium. The high density makes the transmission measurement difficult. The plutonium content of each sample was determined by calorimetry and a gamma isotopic determination; for these samples the content should be accurate to 2%. Figure 3 shows the assay results of the SSC samples. The lump correction for these samples is large (as much as 60%). The SGS assay of these samples is 19% low while the NCC is 25% high. This indicates that the advanced SGS assay technique still has some bias although the bias is substantially reduced. It is interesting to note that if the average of the SGS and the NCC is used, it agrees with the reference values to 4% with an RSD of 5%. This follows the expected trend; SGS will assay low and the NCC will assay high. Consequently the combination brackets the true answer.

The fourth set of samples contains salts generated from the MSE process, in which americium and other impurities are extracted from the plutonium into the salts. The Am/Pu ratio in this salt is quite high (up to 5%) and the americium was physically separated from the plutonium before blending. We determined the plutonium content of these items by pulverizing them, removing metal lumps, blending the rest for an extended time, then taking multiple samples and analyzing them destructively. Chemical analysis was performed on different parts of the sample, and the agreement is between 1.5% and 2%. We feel that the plutonium content is known to 1.5%. Figure 4 shows the results of the SGS measurement of these samples. The SGS gives an average result 4% low, as compared with the reference values, with an RSD of 2.7%. These samples were measured by the SGS before pulverization; the

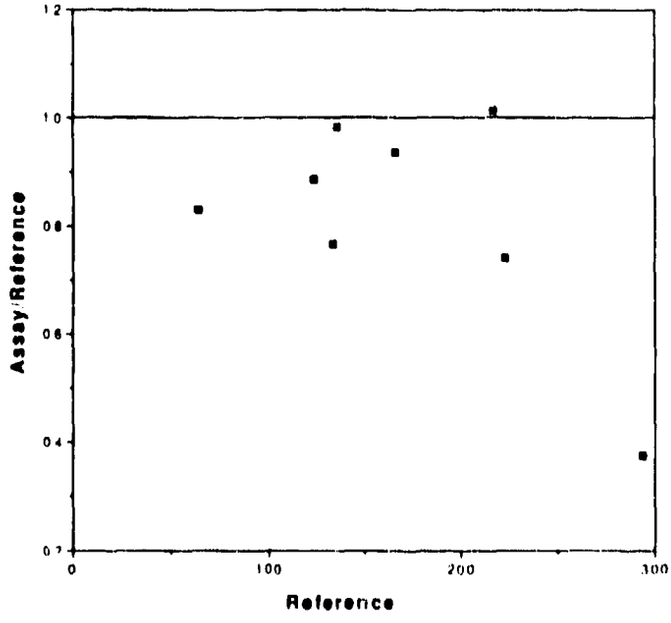

Fig. 3. Assay accuracy of the SGS for the SSC samples. These are noxious waste items not well suited for any measurement. They are heterogeneous and lumpy. The uncertainties in the reference values are about 2%.

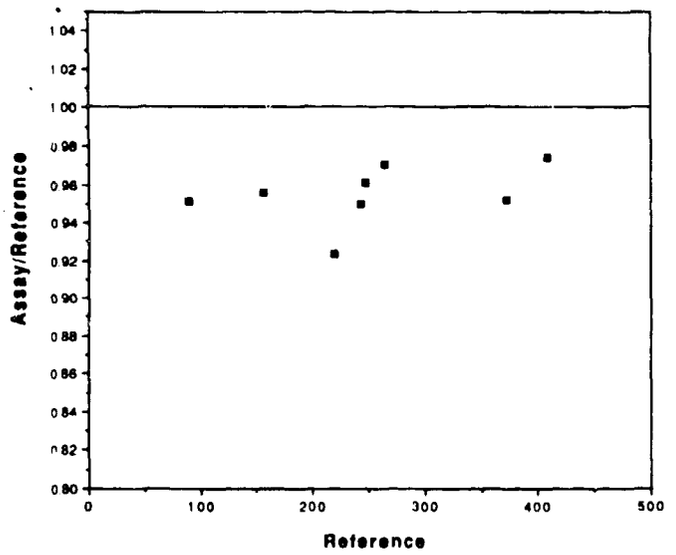

Fig. 4. Assay accuracy of the SGS for homogenized MSE salt scrap. These scrap items are prepared for sampling for chemical analysis to determine plutonium content. After preparation, they are suitable for SGS assay if a valid transmission measurement can be made. The uncertainties in the reference values are approximately 1.5%.

maximum lump correction is 15%. The NCC results are substantially higher than the reference values because of the high americium content; the (α,n) neutrons are the dominant neutrons counted by the NCC. In fact the (α,n) yield is so high that the samples have been measured using the self-interrogation technique.[4] These samples were flagged by the NCC reals-to-totals ratio test (R/T)[5] as being unsuitable for the calibration curve and only the SGS values should be used for the assay result.

The fifth set of samples were fluoride contaminated process sweepings generated at Savannah River Site (SRS). These samples were characterized by calorimetry and gamma isotopics. These measurements were performed at SRS. The LANL calibration was used on the SGS at SRS. The SGS measured the samples with an average bias 0.1% high and an RSD of 1.2%. The NCC had to be recalibrated with these standards because of the substantially different chemical form (fluoride vs oxide). The results of the SGS measurements can be seen in Fig. 5.

CONCLUSION

This study shows that the advanced SGS can measure a wide range of material types and masses. We also showed that the SGS could be calibrated with one standard and still provide good results for a wide variety of samples. Figure 6 shows all of the measured values against the reference values for the various samples measured on the SGS.

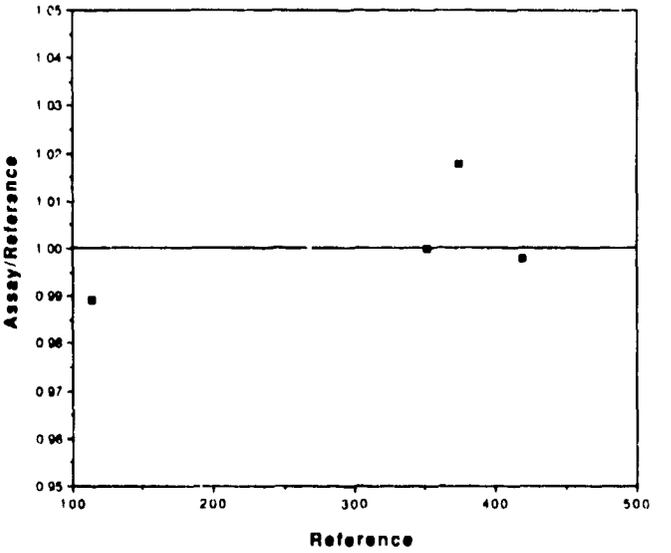

Fig. 5. Assay accuracy of the SGS for the in-plant scrap standards. These scrap items are from a working process. The uncertainties in the reference values are about 2%.

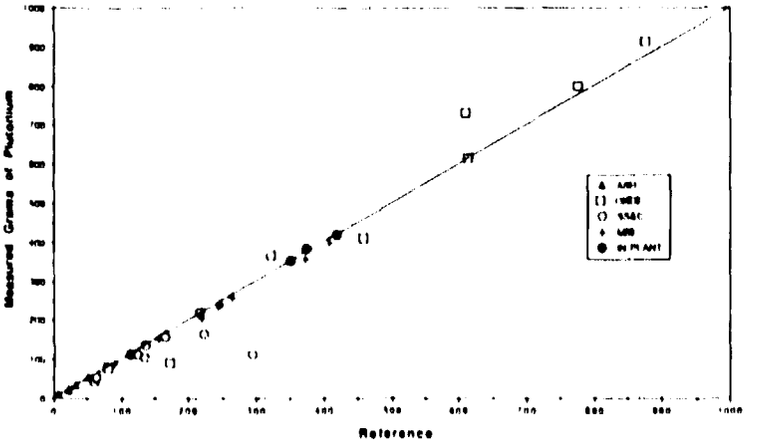

Fig. 6. Assay results of the SGS vs reference values for all of the standards measured.

If the scrap and waste can be separated according to density and material type, the SGS and the NCC can potentially perform assays with biases of less than 5% on some of the most difficult process samples to assay. The SGS and NCC complement each other and, if their results agree to within the uncertainties, we are reasonably confident of the assay result. When the results from both instruments do not agree, we have found it prudent to look at the data and examine the sample more carefully. Often the data from either instrument is obviously flawed. For example, the R/T ratio flag on the NCC may indicate the wrong calibration curve, or that the transmission in the SGS might be too low (<0.001) for a segment of the sample. Obviously, these results cannot be extrapolated to other sample types. These data demonstrate the need to evaluate each scrap and waste stream for measurement accuracy.

Our current efforts on the advanced segmented gamma-ray scanner are focused on improving the lump correction factor technique. We are also exploring the possibilities of making these measurements on SNM materials other than plutonium.

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