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PERFORMANCE OF AN ADVANCED LUMP CORRECTION ALGORITHM FOR GAMMA-RAY ASSAYS OF PLUTONIUM*

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ABSTRACT

The results of an experimental study to evaluate the performance of an advanced lump correction algorithm for gamma-ray assays of plutonium is presented. The algorithm is applied to correct segmented gamma scanner (SGS) and tomographic gamma scanner (TGS) assays of plutonium samples in 55-gal. drums containing heterogeneous matrices. The relative ability of the SGS and TGS to separate matrix and lump effects is examined, and a technique to detect gross heterogeneity in SGS assays is presented.

INTRODUCTION

Self-attenuation of gamma rays by emitting material in the form of small particles or lumps can cause severe bias in gamma-ray assays of special nuclear material. The measured gamma-ray transmissions that are used to correct for sample attenuation are sensitive to attenuation by the sample bulk and do not provide an adequate correction for self-attenuation of the gamma rays by the lumps. In plutonium assays, metallic lumps as small as ~1 mm in diameter can cause significant bias in the assay. In most gamma-ray assays, metal plutonium lumps above ~1 cm in diameter can be considered infinitely thick, resulting in the saturation of the assay results (that is, the results do not change with increasing plutonium mass). The presence of lumps can be detected by examining the variation in assay results as a function of energy, which depends on both the composition of the lumps and the distribution of lump sizes within the sample.

We have developed a lump-correction algorithm that exploits the energy variation of assay results that occurs for lumpy samples.[1] Parameters that describe the effect of lump-size distributions are determined in the fit along with the corrected mass of SNM. The effective atomic number of the emitting material can also be determined in the fit if it is not known.

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Preliminary testing of the algorithm with both simulated and experimental segmented gamma-ray scanner (SGS) assays revealed that additional sources of bias such as matrix inhomogeneities could also give rise to energy variations in the assay result, causing the lump correction algorithm to perform poorly. With the SGS, bias due to source and matrix inhomogeneities is not severe in low-density matrices. However, in medium to high-density matrices, the bias can be significant (see Fig. 6 in Ref. 1). Also, unlike the lump effect, there is no distinct trend in the assay results with energy.

The overall bias due to the combination of lumps and inhomogeneities is multiplicative. Because of the complexity of the variation of assay results due to inhomogeneities, the two bias components cannot easily be separated using SGS data. However, accurate corrections for self-attenuation should be possible using the tomographic gamma-ray scanner (TGS) because it potentially provides a complete correction for heterogeneity in the matrix and in the distribution of emitted material.[2]

To test this hypothesis, we assembled a set of drums containing a variety of matrix types, including both uniform and heterogeneous matrices. A set of four SGS standards with ^{239}Pu masses ranging from 10 to 100 g were loaded in various configurations in the drum to simulate distributed sources and to provide calibration points for TGS and SGS assays. A set of five "2-g" plutonium samples consisting of plutonium oxide powder in vials, approximately 1.5 in. long by 0.5 in. in diameter, were used to simulate the effect of lumps. The total ^{239}Pu content in the vials used in the study was known to be 8.24 g.

DATA ACQUISITION AND ANALYSIS

The test drums were assayed using the prototype TGS, jointly developed by NIS-5 and NIS-6 at Los Alamos. The WIN_TGS[3] code was used to acquire transmission and emission data separately using a two-pass continuous tomographic scanning protocol, described in Refs. 2 and 4. In each pass, 2250 measurements are made, from which roughly 1600 image parameters are determined. Single-pass scan times were

varied from 25 min to 4 h to determine the effect of counting statistics on assay precision.

A 30 mCi, ^{75}Se source was used to determine transmissions at 136, 208, and 401 keV. Emission data were acquired for the 129-, 203-, 345-, and 414-keV lines of ^{239}Pu. A rate loss source (^{109}Cd, 88 keV) was used to correct both the transmission and emission measurements.

The data was analyzed using the TCNDA code developed at Los Alamos.[4,5] The analysis of TGS data involves four steps:

1. Interpolation of measured transmissions to determine transmissions at the assay energies,
2. Reconstruction of the attenuation coefficient distribution for assay energy from the interpolated transmissions,
3. Reconstruction of transmission-corrected emission images for each assay energy,
4. Post-processing, including the determination of total mass, lump corrections, and precision estimates.

Experimental TGS data were also collapsed to create SGS data for the comparative analysis.

CALIBRATION

The TGS was calibrated using a set of repeated assays of a 30-g plutonium SGS standard in a drum containing polyethylene shavings. The position of the sample within the drum was varied, and the assay results were averaged to determine a calibration constant for each energy. The results of three additional assays of the 30-g source are shown in Fig. 1 for the TGS (squares) and the SGS (circles). Two additional TGS assay results for the same sample in a drum with a nonuniform matrix are shown (triangles) to illustrate the ability of the TGS to correct for matrix heterogeneity. The mass of the standard was ~28 g of ^{239}Pu.

LUMP CORRECTION EXPERIMENTS

Assays of a single ~2-g PuO_2 vial ir an empty drum were used to determine the level of self-attenuation in the "2-g" samples. The results of a single TGS assay (squares) and two SGS assays (diamonds) are shown in Fig. 2. The average variation of assay mass with energy is shown by the solid line. The lump-correction algorithm was applied to the average assay result yielding a correction factor of ~1.33 at 414 keV.

The complete set of "2-g" samples was randomly distributed within drums containing medium-density

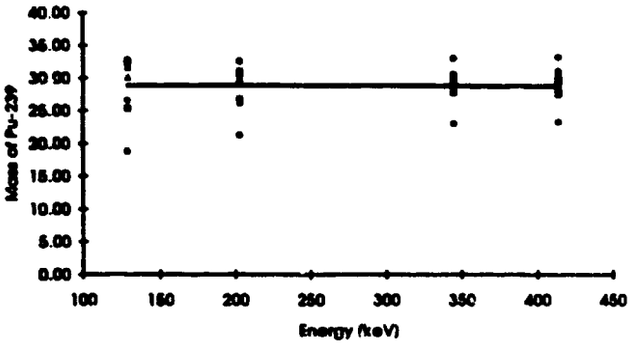

Fig 1. SGS and TGS assays of a 30-g SGS plutonium standard in 55-gal. drums containing uniform and heterogeneous matrices.

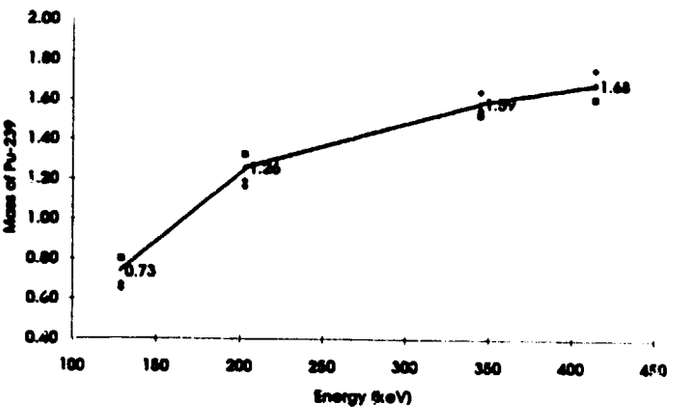

Fig. 2. TGS and SGS assay results for a 2-g plutonium oxide vial in an empty drum.

matrices with a relatively large degree of heterogeneity. TGS assay results for five cases are shown in Fig. 3. The expected variation of assay mass with energy is shown by the solid line. The line was determined by scaling the average variation of assay mass with energy determined previously from the empty-drum assays of the single PuO_2 vial. The scaling factor was obtained by first normalizing the empty-drum curve to the 414-keV mass and then scaling by a factor of 8.24/1.33.

The TGS assays were independently analyzed using the lump-correction algorithm. Both the effective lump size and the effective atomic number of the lumps were determined in each fit. The average correction factor for the 414-keV mass was found to be 1.35, differing by less than 2% from the correction factor determined for the empty drum. The average corrected mass value was 7.94 g (±) 7%, which is 3.64% lower than the known mass of 8.24 g. The standard deviation in the distribution of corrected masses was ~13%. The average reduced chi-

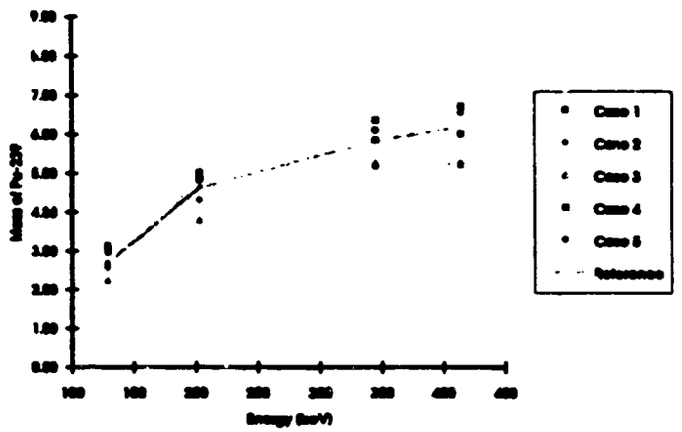

Fig. 3. TGS assays of the plutonium-oxide vials in heterogeneous drums.

squared value determined by the correction algorithm was ~1.2, with 1 degree of freedom.

SGS assay results for the five cases are shown in Fig. 4 along with the expected variation of assay mass. Two of the cases depart significantly in magnitude and shape from the expected trend. In these cases, large chi-squared values determined by the lump-correction algorithm indicate that the observed variation in assay mass cannot be attributed to lumps.

Because matrix and lump effects are multiplicative, the residual bias due to matrix heterogeneity can be determined if independent knowledge of the variation of mass due to lump effects is known. In the cases examined, the residual bias can be determined by dividing each mass value determined in the assay by the corresponding average mass value determined for the empty-drum case. For the TGS assays, the residual bias due to matrix heterogeneity is shown in Fig. 5. Note that there is a considerable amount of scatter for the lower energies; however, the average residual bias indicated by the solid line is less than 5% over the entire range of energies, indicating the overall effect of matrix heterogeneity is small.

In SGS assays, the residual bias is considerable for two of the cases examined, particularly for the 129-keV line shown in Fig. 6. In both cases, the TGS emission images revealed that the vials were placed toward the edge of the drum. Experience with simulated assays has shown that a decreasing trend of SGS mass with energy is typical for cases in which the emitting material is located near the edge of the drum in a dense or heterogeneous matrix.

CONCLUSIONS

This comparative study between TGS and SGS assays using lumpy samples provides preliminary experimental evidence that bias due to source self-attenuation can be obscured by matrix heterogeneity in SGS assays. Because the bias due to matrix effects in TGS assays is small, the TGS can more accurately correct for the bias caused by lumps. With SGS, the observation of increasing assay mass with energy does not imply the presence of lumps. Failure of the goodness-of-fit test provided by the least-squares lump-correction algorithm can serve as a practical way to detect gross heterogeneities in the matrix.

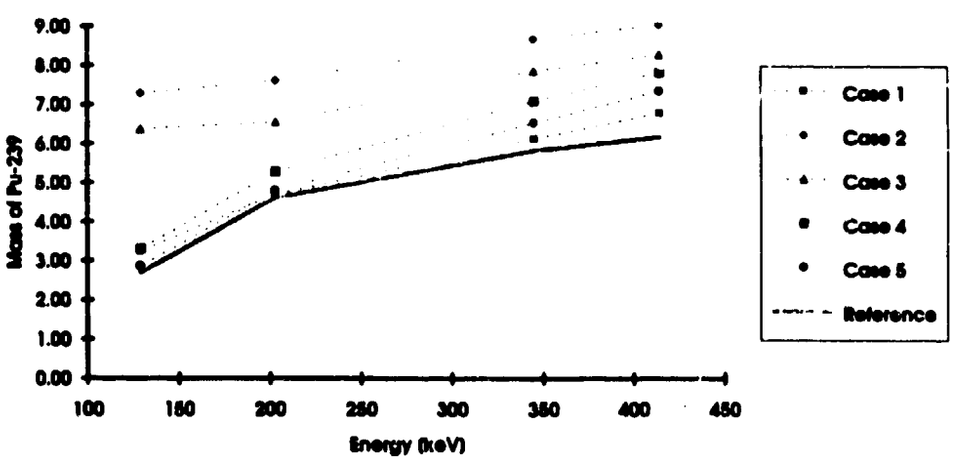

Fig. 4. SGS assays of the plutonium-oxide vials in heterogeneous drums.

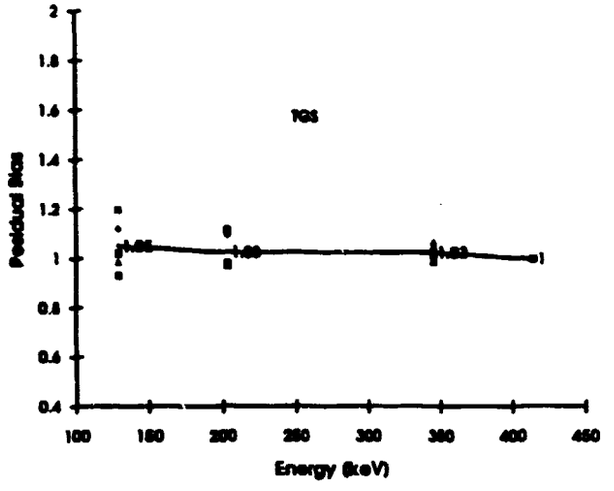

Fig. 5. Residual bias due to matrix effects for TGS.

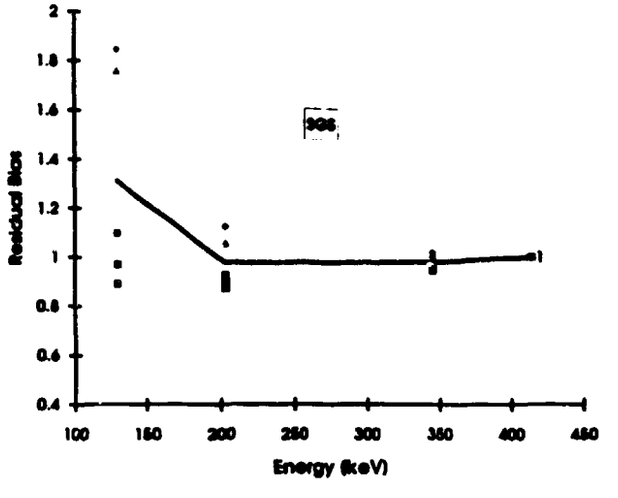

Fig. 6. Residual bias due to matrix effects for SGS.

A practical application of the goodness-of-fit test is in hybrid gamma-ray systems that are capable of performing both SGS and TGS assays. When throughput is important, the SGS mode can be utilized. SGS assays that fail the goodness-of-fit test following the application of the lump correction algorithm can be assayed using the TGS. In facilities where the majority of drums contains benign matrices that are suitable for assay with the SGS, this technique should result in a considerable increase in throughput without compromising the accuracy of the assays.

ACKNOWLEDGMENTS

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REFERENCES

1. T. H. Prettyman, J. K. Sprinkle, and G. A. Sheppard, "A Weighted Least-Squares Lump Correction Algorithm for Transmission-Corrected Gamma-Ray Nondestructive Assay," *Nucl. Mater. Manage.* **XXII** (Proc. Issue), 682-690 (1993).

2. R. J. Estep, T. H. Prettyman, and G. A. Sheppard, "Tomographic Gamma Scanning (TGS) to Measure Inhomogeneous Nuclear Material Matrices from Future Fuel Cycles," Los Alamos National Laboratory document LA-UR-93-1637 (May 1993).

3. R. J. Estep and J. T. Cavender, "The WIN-TGS Software Package for Tomographic Gamma Scanner Systems," presented at Institute of Nuclear Materials Management meeting, Naples, Florida, July 17-20, 1994.

4. T. H. Prettyman, R. J. Estep, and G. A. Sheppard, "Development of a Tomographic Instrument for Gamma-Ray Nondestructive Assay," *Trans. Am. Nucl. Soc.* **69**, 182-184 (1993).

5. T. H. Prettyman, R. A. Cole, R. J. Estep, and G. A. Sheppard, "A Maximum-Likelihood Reconstruction Algorithm for Tomographic Gamma-Ray Nondestructive Assay," Los Alamos National Laboratory document LA-UR-94-1650 (May 1994).