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An innovative method for extracting isotopic information from low-resolution gamma spectra

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A method is described for the extraction of isotopic information from attenuated gamma ray spectra using the gross-count material basis set (GC-MBS) model. This method solves for the isotopic composition of an unknown mixture of isotopes attenuated through an absorber of unknown material. For binary isotopic combinations the problem is nonlinear in only one variable and is easily solved using standard line optimization techniques. Results are presented for NaI spectrum analyses of various binary combinations of enriched uranium, depleted uranium, low burnup Pu, ^{137}Cs , and ^{133}Ba attenuated through a suite of absorbers ranging in Z from polyethylene through lead. The GC-MBS method results are compared to those computed using ordinary response function fitting and with a simple net peak area method. The GC-MBS method was found to be significantly more accurate than the other methods over the range of absorbers and isotopic blends studied.

1. Introduction

It was recently shown[1] that a logarithmic gamma-ray response function analysis technique used in tomographic assays[2,3] can be modified to solve for the isotopic composition of an unknown radioactive source attenuated by an absorber of unknown composition, a problem that previously required extensive radiation transport computations[4] to solve. Because this new approach – termed the gross-count material basis set (GC-MBS) method – works with NaI, CdZnTe, or other low-resolution detectors, it has potential application in portable hand-held radiation monitors.

In this report we evaluate the accuracy of the GC-MBS method in analyzing NaI spectra for the isotope fractions of various binary mixtures of HEU (highly enriched uranium), DU (depleted uranium), low-burnup Pu, ^{137}Cs , and ^{133}Ba transmitted through different absorbers. We compare the GC-MBS results with those obtained using conventional response-function fitting and a simple net-area method.

2. The GC-MBS Method for Isotopic Analysis

For an isotopic composition represented by the unattenuated spectrum $\mathbf{c}_{\text{ref}}$ of a reference source of mass mass_{ref} , the GC-MBS method models the observed log-ratio spectrum $\mathbf{v}$ from an unknown source as a system of n_e equations in n_z+1 unknowns,

$$\mathbf{v} = \mathbf{U} \rho \quad , \quad (1)$$

where n_e is the number of usable channels in the spectrum and n_z is the number of basis materials in the MBS representation. The elements of the vector $\mathbf{v}$ are calculated from the individual channel counts c_e of the unknown's spectrum and those ($c_{\text{ref},e}$) of the reference spectrum as

$$v_e = -\ln(c_e/c_{\text{ref},e}) \quad . \quad (2)$$

The first n_z columns of the matrix $\mathbf{U}$ are the log-transmission spectra $\mathbf{v}_z$ ($U_{ez} = v_{z,e} = -\ln\{c_{z,e}/c_{\text{ref},e}\}$, $z = 0, \dots, n_z-1$) for attenuation-distorted spectra $\mathbf{c}_z$ of the reference source measured through each of the n_z basis materials. The last column of $\mathbf{U}$ is given by $U_{e,nz} = 1$ for all e .

The solution vector $\rho = [\rho_0 \dots \rho_{n_z-1} \ x]$ gives both the absorber composition in the MBS representation (ρ_z is the amount of material z) and the mass-dependent term x . The unknown source mass (mass) is given relative to the reference source mass by

$$\text{mass} = \text{mass}_{\text{ref}} \cdot \exp(-x) \quad . \quad (3)$$

Equation (1) is applied to a mixture of two isotopes* A and B with isotope fractions y_A and $y_B (= 1 - y_A)$ by blending the reference and basis spectra for pure A and B, i.e.,

$$\mathbf{c}_{\text{ref}} = y_A \cdot \mathbf{c}_{\text{ref},A} + (1 - y_A) \cdot \mathbf{c}_{\text{ref},B} \quad \text{and}$$

$$\mathbf{c}_z = y_A \cdot \mathbf{c}_{z,A} + (1 - y_A) \cdot \mathbf{c}_{z,B} \quad . \quad (4)$$

The nonlinear method used here to solve for an unknown isotope fraction y_A in a binary mixture is to search for the value of y_A that minimizes χ^2 for the linear GC-MBS fit in Eqn. (1). This is a simple line optimization problem that can be solved by any of several search algorithms.

3. Experimental Details and Data Handling

The raw data were acquired using a simple source-absorber-detector geometry. The source was placed along the axis of the detector at a distance of 16.5 cm. The absorbers were placed between the source and detector such that the detector was entirely shielded by the material. The raw data were detected using a 5.04×5.04 cm NaI detector (Bicron model 2M2/2). This data was collected and saved using a PC with the Ortec GammaVision © software package and an Ortec model

* or well-defined blends of isotopes such as HEU

918 multi-channel analyzer card[5]. Spectra were compressed from 4096 channels to 256 channels prior to GC-MBS and ordinary response-function analysis.

The absorbers tested (18 materials plus the unattenuated case) consisted of several thicknesses of five materials and three binary combinations of the five materials. These are listed in Table I, with the five GC-MBS basis materials indicated by asterisks.

Table I. Attenuating Materials

Material	Thicknesses Used (cm)
None*	0
Aluminum	1.27, 2.54, 6.35*
Polyethylene	2.54, 5.08, 10.16*
Cadmium	0.318*, 0.635, 0.953
Carbon Steel	0.635, 1.27*, 2.54
Lead	0.318, 0.635*, 1.27
Mixture 1	10.16 poly.+0.318 cadmium
Mixture 2	1.27 steel + 2.54 poly.
Mixture 3	1.27 aluminum + 0.635 lead

* Used as a basis/reference spectrum in the GC-MBS

Separate 300-s spectra were collected for each of the sources [HEU (100 g), DU (10 kg), low burnup Pu (30 g), ^{137}Cs (10 μCi), and ^{133}Ba (10 μCi)] transmitted through each of the absorbers in Table I. The spectra were normalized to have the same number of counts in the unattenuated case within the GC-MBS fitting region. To generate a series of "unknown" spectra of mixed isotopic composition, we added the pure spectra in the proportions $y_A=0, 0.1, 0.25, 0.5, 0.75, 0.9$ and 1, generating 133 spectra for each source pair studied.

We used the LANL-developed C program GROSSFIT as described in ref. [1] to solve the GC-MBS problem (Eqn. (1)) for each trial value of the primary isotope fraction y_A . A second program, MIXER, generated the blended trial basis spectra (c_{ref} { c_z }) as in Eqn. (4). The C program BINARY automated the nonlinear isotopic analysis for binary mixtures by generating the trial y_A values and alternately spawning the MIXER and GROSSFIT programs. A simple block search algorithm in BINARY finds the y_A that minimizes χ^2 to within ± 0.004 .

The net-area method uses the background-subtracted area under the major peaks or peak groups of isotopes A and B to estimate y_A . For ordinary response function fitting we analyzed the unknown spectra using the computer program ISOTOPIX, a LANL-developed C program that uses the NNLS (non-negative least squares) algorithm[6] to solve the n_e by n_a system of equations

$$c = R a \quad (5)$$

where the columns of the matrix R are the unattenuated reference spectra $c_{\text{ref},i}$ ($i=0, \dots, n_a-1$) for the n_a isotopes

Fig 1. The logarithm of χ^2 as a function of the ^{137}Cs fraction y_{Cs} for various blends of ^{137}Cs and ^{133}Ba transmitted through 1.28 cm of aluminum plus 0.64 cm of lead. The y_{Cs} values that minimized χ^2 in the GC-MBS method are shown in parentheses.

being modeled (in this case $n_a=2$) and the elements a_i of the solution vector a are the amounts of each isotope. These are converted to their corresponding isotope fractions y_i for comparison with the GC-MBS results.

4. Results and Discussion

Figure 1 shows χ^2 for the CG-MBS fit in Eqn. (1) as a function of the ^{137}Cs fraction y_{Cs} for $^{137}\text{Cs}/^{133}\text{Ba}$ blends with $y_{\text{Cs}} = 0, 0.1, 0.25, 0.5, 0.75, 0.9$, and 1.0 transmitted through an absorber of 1.28 cm of aluminum plus 0.64 cm of lead. As indicated in parentheses in the figure, χ^2 is minimized at $y_{\text{Cs}} = 0.008, 0.108, 0.252, 0.500, 0.748, 0.888$, and 1, respectively, in excellent agreement with the correct fractions. In contrast, for the same case the net-area method predicted $y_{\text{Cs}} = 0, 0.257, 0.53, 0.766, 0.893, 0.944$, and 0.97, while the ISOTOPIX code predicted $y_{\text{Cs}} = 0.094, 0.306, 0.516, 0.722, 0.843, 0.895$, and 0.924. The GC-MBS method gave similar improvements in accuracy with the other absorbers in the series. This is illustrated in Fig. 2, which shows the absolute error in y_{Cs} when estimated by the GC-MBS method versus the error in the ISOTOPIX estimate, for transmission of the seven isotopic blends of $^{137}\text{Cs}/^{133}\text{Ba}$ through the nineteen absorbers. Overall, for $^{137}\text{Cs}/^{133}\text{Ba}$ mixtures the GC-MBS method exhibited an rms error of only 3.8%, compared with 16% for the net-area and 10% for the ISOTOPIX results.

Fig. 2. Absolute error in the GC-MBS estimate of the ^{137}Cs fraction in blends of ^{137}Cs and ^{133}Ba as a function of the error in the ISOTOPIX estimate. The GC-MBS results within the diagonals have less error than the ISOTOPIX estimate.

Intuitively, we expect that the largest improvement in accuracy with the GC-MBS approach will occur for isotope blends in which the major peak groups overlap and are thus difficult to resolve, and which also have fairly complex spectra individually. Complex spectra (i.e., those with multiple energies in broad groups) provide more useful information for the GC-MBS method to solve the absorber composition problem than do simple spectra, such as those from ^{137}Cs . For these reasons, the Pu/ ^{133}Ba blends showed the best improvement when using the GC-MBS method. This is illustrated in Fig. 3, which shows the error in the GC-MBS estimate of the Pu fraction in blends of low-burnup Pu and ^{133}Ba as a function of the error in the ISOTOPIX estimate.

The average and rms errors observed in our net-area, ISOTOPIX and GC-MBS analyses are summarized in Table II for five binary source combinations and for all measurements combined. In each case the rms error is significantly less with the GC-MBS method than with either of the other methods and is within tolerable limits for many potential applications. The average GC-MBS errors are less than 2% in all cases.

Table II. Error in Estimated Isotopic Fractions

Isotope Blend	Average* $\pm$ RMS Error for:		
	Net Area Method	ISOTOPIX Code	GC-MBS Method
HEU-DU	.15 $\pm$.27	.31 $\pm$.41	.01 $\pm$.11
Pu- ^{137}Cs	-.04 $\pm$.09	.02 $\pm$.08	0 $\pm$.016
Pu- ^{133}Ba	-.18 $\pm$.30	.02 $\pm$.21	0 $\pm$.038
Pu/HEU	.25 $\pm$.48	.12 $\pm$.24	-.015 $\pm$.068
^{137}Ba - ^{137}Cs	-.06 $\pm$.16	-.01 $\pm$.10	.01 $\pm$.038
All cases	.02 $\pm$.29	.09 $\pm$.24	.001 $\pm$.06

* Average of the true-estimated primary isotope fraction for 133 cases (19 absorbers times 7 isotopic fractions).

The net-area method showed large average and rms errors in analyzing the Pu/HEU blends. This is a difficult case for the net-area method because the main peak groups are not resolved in NaI spectra. Where the peak groups were resolved, the net-area method worked about as well as the ISOTOPIX fit, and for the HEU/DU mixture it was significantly more accurate (although still less accurate than the GC-MBS method by a factor of 2.5).

Fig. 3. Absolute error in the GC-MBS estimate of the Pu fraction in blends of low-burnup Pu and ^{133}Ba as a function of the error in the ISOTOPIX estimate.

For both the ISOTOPIX and GC-MBS methods, HEU/DU proved the most difficult case for measurement of the isotope fraction. This is apparent in Fig. 4, which shows the error in y_{HEU} estimated by the GC-MBS method versus the error in the ISOTOPIX estimate for all absorbers and blends. What we observe is that the 11% rms error distribution for the GC-MBS results does not appear to be gaussian, but seems instead to be composed of a narrower distribution of approx. 5% rms error with an unusually large number of outliers. Most of the outliers can be identified with the most attenuating absorbers (e.g., 1.27 cm of lead), which the 186-keV group of gamma rays in HEU cannot penetrate. Even so, the 11% rms error distribution for the GC-MBS method is a significant improvement over the 27% and 41% rms error distributions for the net-area and ISOTOPIX approaches.

Fig. 4. Absolute error in the GC-MBS estimate of the HEU fraction in blends of HEU and DU as a function of the error in the ISOTOPIX estimate.

Acknowledgments

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