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A SYSTEM FOR THE ANALYSIS OF GAMMA-RAY SPECTRA BY THE METHOD OF SIMULTANEOUS EQUATIONS

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ABSTRACT

A complete system for the quantitative separation of complex gamma-ray spectra into their component nuclides was developed and programmed in FORTRAN IV for the IBM 360/65. This system was developed to analyze routine data generated by two multichannel analyzers (three crystal systems) at the Savannah River Laboratory. The system also maintains a current library of standard spectra. The method of simultaneous linear equations is employed.

CONTENTS

INTRODUCTION

One of the responsibilities of the Savannah River Laboratory is the routine sampling of the environs both on and around the Savannah River Plant site. The use of NaI(Tl) scintillation crystals with multichannel analyzers to analyze these samples for levels of activity from less than 1 to 10,000 counts/minute has resulted in considerable savings in time, labor, and cost. In the use of scintillation spectrometers, one of the difficult problems is the quantitative separation of complex gamma-ray spectra into their component nuclides. If the problem is treated as one of unknown nuclides and unknown intensities, then the problem is extremely difficult and an attempt at solution has been made by using stepwise multiple regression.[1] If however, the qualitative contents of the sample are known, then the solution to the problem is much easier. This assumption was made in the method of simultaneous linear equations,[2] which is used in the programs described in this report. Other methods, such as spectrum stripping,[1] least squares,[3] and mathematical programming,[4] are applicable to the problem, but the method of simultaneous linear equations best meets the objectives of the Savannah River Laboratory.

SUMMARY

A system of computer programs called GAMSPEC was written in FORTRAN IV for use on the IBM 360/65 and have been successfully applied to the analysis of combinations of the following nuclides:

^{7}Be	^{60}Co	^{134}Cs
^{24}Na	^{65}Zn	^{137}Cs
^{40}K	^{85}Sr	^{140}Ba
^{51}Cr	^{95}Zr	^{141}Ce
^{54}Mn	^{103}Ru	^{144}Ce
^{57}Co	^{106}Ru	^{226}Ra
^{58}Co	^{125}Sb	^{233}Pa
^{59}Fe	^{131}I	^{239}Np

The following table shows a comparison between the results of GAMSPEC for a standard mixture of three nuclides and the known values in the mixture.

Nuclide	Known Values, d/m	Measured by GAMSPEC, d/m	Ratio Measured/Known
^{144}Ce	1.00	1.03	1.03
^{137}Cs	5.20	5.31	1.02
^{60}Co	5.00	5.11	1.02

The accuracy of these results is representative of those obtained with GAMSPEC. In general, the accuracy for a specific sample is least when the number of nuclides to be resolved is greatest and when photopeak channel groupings are closest together.

DISCUSSION

DESCRIPTION OF THE CRYSTAL SYSTEMS

The GAMSPEC system is applied to the output from three crystal systems. Two of these systems are 3" x 3" NaI(Tl) crystals encased in stainless steel with 4" of lead shielding. Each of these is connected to a 200-channel portion of a 400-channel RIDL transistorized analyzer. The other system is a 9" x 9" NaI(Tl) crystal with a 3" diameter x 6" deep well, which is covered by aluminum; the remainder of the crystal is encased in stainless steel with 4" of lead shielding. This system, which is from 5 to 10 times more sensitive than the 3" x 3" systems, is connected to a 400-channel RIDL transistorized analyzer with automatic sample changer.

METHOD OF CALCULATION

The method of calculation is based on solving a system of simultaneous linear equations, which are set up on the assumption that the qualitative contents of the sample are known. It is also assumed that every sample spectrum is a linear combination of component (library) spectra, except for random statistical errors.

In using the method of simultaneous linear equations, a 200-channel input spectrum is effectively reduced to a K channel spectrum where K is the number of nuclides in the sample. Each of these K reduced channels consists of a predefined set of analyzer channels, selected around a photopeak of the nuclide of interest. These predefined sets of channels are called photopeak channel groupings. The number of channels per photopeak channel

grouping is somewhat arbitrary and is chosen to minimize overlap between two groupings and to accommodate the increased width of the photopeak with increasing energy.

Consider a sample containing K gamma-emitting nuclides, X_1, X_2, ... X_K. Then the total observed counts in the photopeak channel grouping for X_i would be

$$T_i = F_{i1}N_1 + \ldots + F_{ij}N_j + \ldots + F_{iK}N_K + B_i$$

where

T_i = total observed counts in the photopeak channel grouping for X_i

N_j = counts due to X_j that appear in the photopeak channel grouping for X_j

F_{ij} = fraction of N_j that appears in the photopeak channel grouping for X_i

B_i = background in photopeak channel grouping for X_i

Therefore, we have the system,

$$\sum_{j=1}^{K} F_{ij}N_j = T_i - B_i, \qquad (i = 1, 2, 3, \ldots, K) \qquad (1)$$

to solve for N_j, where the T_i are determined by integrating the observed sample count over the predetermined photopeak channel grouping, and the B_i are similarly determined from a precounted background. The F_{ij} are obtained from the spectra library and are defined so that the F_{jj} are unity.

In matrix notation, equation (1) can be written as

$$FN = C$$

where $F = (F_{ij})$; $N = (N_j)$; and $C = (T_i - B_i)$. The solution of this set of K equations in K unknowns is given by $N = F^{-1}C$. When weighted by the appropriate time of count, sample volume, decay constant, and conversion factors, N gives the amount of each radionuclide present in the sample. This procedure assumes that the calibration constants determined previously are valid at the time of the sample count and that no nuclide is present in the sample that has not been previously identified.

Example

Consider a sample with two nuclides, X_1 and X_2. Let the predetermined photopeak channel groupings for X_1 and X_2 be channels 34-38 and channels 62-70, respectively. The decay constant, conversion factors, sample volume, and time of count are all unity.

Channel	Count	Background	
34-38	$T_1 = \sum_{34}^{38} = 1020$	$B_1 = \sum_{34}^{38} = 20$	$F_{11} = 1.0$ $F_{21} = .01$
62-70	$T_2 = \sum_{62}^{70} = 2050$	$B_2 = \sum_{62}^{70} = 50$	$F_{12} = .20$ $F_{22} = 1.0$

Then from equation (1),

$$1020 - 20 = N_1 + .2N_2$$
$$2050 - 50 = .01\,N_1 + N_2$$
$$\begin{pmatrix} 1 & .2 \\ .01 & 1 \end{pmatrix}\begin{pmatrix} N_1 \\ N_2 \end{pmatrix} = \begin{pmatrix} 1000 \\ 2000 \end{pmatrix}$$
$$\begin{pmatrix} N_1 \\ N_2 \end{pmatrix} = \begin{pmatrix} 601 \\ 1994 \end{pmatrix}$$

Therefore, there are 601 units of nuclide X_1 in the sample and 1994 units of nuclide X_2.

DESCRIPTION OF THE COMPUTER SYSTEM

GAMSPEC was written in FORTRAN IV for a 256K IBM 360/65 with 7 tapes and a 2314 direct access storage device. It is set up to handle 42 library nuclides and up to 20 nuclides per sample. The system consists of four major computer routines, BASIC, CHSUM, PROFRM, and CALC, which can be utilized as separate programs or as a continuous machine run. The system can take input data in through any one of its major routines and update that part of the system, which the major routine controls. The overall system flow is shown in Figure 1.

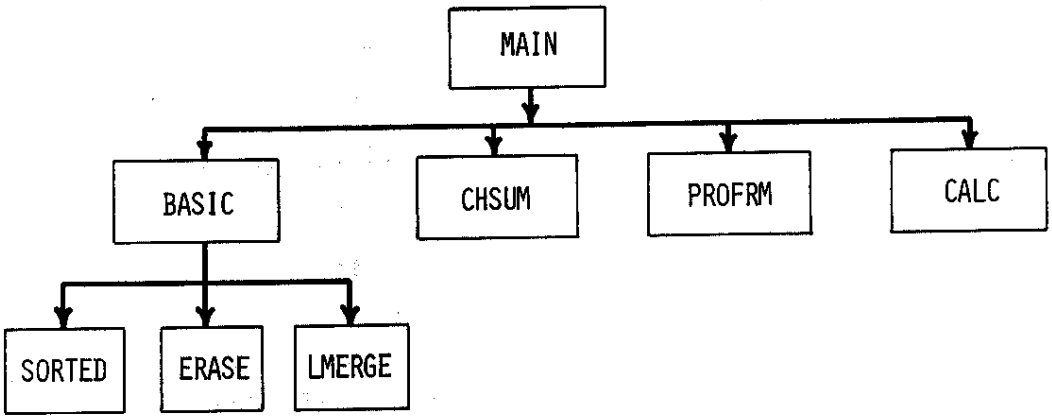

FIG. 1 SYSTEM FLOW TO MAJOR ROUTINES

The first routine, BASIC, sets up or updates a basic calibration library of "mono-nuclidic" gamma-ray spectra. It maintains a series of records on a master library tape, each record being a particular calibration count of a specified nuclide. BASIC can accept new calibration counts into the system, sort them, delete specified counts for a particular nuclide from the master, and merge the master and the new calibration data with appropriate output.

The second routine, CHSUM, determines the F_{ij}, described under the method of solution, by calculating the fractional "scatter" contributions from the assigned photopeak channel grouping of each library nuclide into the assigned photopeak channel grouping of every other library nuclide. Provision is made for several different groups of photopeak channel groupings for a given geometry whenever one is insufficient. CHSUM then sets up a library of these fractional contributions for use in the next routine, PROFRM. CHSUM also calculates a 1σ error bound on these fractional contributions to be used in routine CALC for error determination.

The third routine, PROFRM, combines up to 20 radionuclides from the CHSUM library into various "nuclide programs", each composed of nuclides characteristic of a particular type sample and/or location. The matrices of fractional contributions are inverted and stored in a PROFRM library to be used by routine CALC.

The fourth routine, CALC, performs the actual separation, using the method of simultaneous linear equations previously described and the output generated by routines, BASIC, CHSUM, and PROFRM, as calculation constants. Accuracy bounds, δN, on the calculated results are determined from

$$\delta N = F^{-1} [(\delta C) - (\delta F)N] \qquad (2)$$

where δC is the error vector for C, and δF is the error matrix for F. This relation is derived as follows:

Although the set of equations solved is

$$FN = C$$

the true values of the coefficients are $F + \delta F$, the true values of the right-hand side are $C + \delta C$, and the true values of the unknowns are $N + \delta N$. Hence

$$(F + \delta F)(N + \delta N) = C + \delta C$$

but with $FN = C$ and $(\delta F)(\delta N)$ considered negligible,

$$(\delta F)N + F(\delta N) = \delta C$$

or

$$\delta N = F^{-1}[(\delta C) - (\delta F)N]$$

as stated in equation (2).

For a given sample, δF and δC are not known, but only the estimated standard deviations in F and C. The estimated standard deviations for F are determined in CHSUM from counting replicate samples for each nuclide, while those for C are determined from the assumption that the sample data follow a Poisson distribution. Therefore,

$$\delta F \approx 2\sigma_F$$

$$\delta C \approx 2\sigma_C = 2\sqrt{T + B}$$

are used in equation (2) to calculate a 95% confidence band on the calculated nuclide intensities.

A set of operating instructions for GAMSPEC are given in the appendix.

APPENDIX

OPERATING INSTRUCTIONS FOR GAMSPEC

The major subroutines, BASIC, CHSUM, PROFRM, and CALC, are numbered 1, 2, 3, and 4, respectively. They may be called in increasing sequence with user-determined beginning and ending routines.

Card 1	Column	Description
	1	Number of major routines requested Maximum - 4
	2	Identification of first routine used May be BASIC, CHSUM, PROFRM, CALC
	3	Identification of second routine used May be CHSUM, PROFRM, CALC
	4	Identification of third routine used May be PROFRM, CALC
	5	Identification of fourth routine used CALC

Example

The card 2 3 4 would indicate that two routines are desired; these routines are PROFRM and CALC.

Switch Settings

Card 2	Column	Subroutine	Description
	1	PROFRM	1 - Merge old and new tapes in PROFRM
			0 - Do not merge tapes
	2	BASIC	1 - Print old BASIC tape in ERASE without processing
			0 - Do not print tape
	3	CALC	1 - +, 0, - right hand members
			0 - +, 0 right hand members
	4	BASIC	1 - Print output from SORTED
			0 - Do not print output

Card 2	Column	Subroutine	Description
	5	BASIC	1 - Print updated tape from ERASE and/or LMERGE
			0 - Do not print updated tape from ERASE and/or LMERGE
	6	CHSUM	1 - Print scatter fractions for each sample in CHSUM
			0 - Do not print scatter fractions for each sample

OPERATING INSTRUCTIONS FOR BASIC

General

The BASIC subroutine serves as the basic input subroutine for the Gamma Spectroscopy Calibration programs. It accepts a BCD calibration tape (Data Set 11) prepared by the IBM 1401, sorts the data, erases selected data from the old master (Data Set 12), and merges the new calibration data with the old master to give an updated tape (Data Set 16).

BASIC calls a combination of the following three subroutines:

1. SORTED accepts the BCD calibration tape and outputs a tape of the data sorted by geometry and nuclide (Data Set 22).

2. ERASE accepts the old master tape (Data Set 12), erases blocks of data as called for by control cards, and writes out the retained data (Data Set 3). This is the only subroutine of BASIC that requires control cards.

3. LMERGE accepts two data sets (Data Sets 22; 12 or 3) and writes a merged tape (Data Set 16). In the production runs, the usual procedure will be to run SORTED, then LMERGE, which will have the effect of updating the master tape of basic calibration data.

BASIC Data Sets

Number	Device	Subroutine	Purpose
2	DA	SORTED, ERASE	Scratch
3	DA	SORTED, LMERGE	Scratch
8	DA	SORTED	Scratch
9	DA	SORTED	Scratch
11	7-Trk	SORTED	Input of new calibration data
12	9-Trk	ERASE, LMERGE	Current master library Previous output from BASIC
13	DA	SORTED	Scratch
16	9-Trk	LMERGE	New master library New output from BASIC
22	9-Trk	SORTED, LMERGE	Sorted calibration data Output from SORTED Input to LMERGE

BASIC Control Cards

Card 1 Column	Description
1	Number of routines requested May be 1, 2, or 3
2-4	Code numbers of desired routines - 1 - SORTED 2 - ERASE 3 - LMERGE

Example

2 1 3 would indicate that two routines are desired: these routines are SORTED and LMERGE.

SORTED Control Cards (None required)

ERASE Control Cards

K - number of geometries having nuclides to be erased.

Card(s) 1 - K	Column	Description
	1-2	Geometry in which erasures occur
	3-4	N - Number of nuclides to be erased
		N - 0 if all nuclides are to be erased for this geometry
	5-6	Number of the first nuclide to be erased (right-justified)
	7-8	M(1) - Number of samples to be erased for the nuclide
		M(1) - 0 if all samples are to be erased for this nuclide
	9-10	Same as 5-8 for second nuclide to be erased
	11-12	
	⋮	

Example

_ 2 _ 4 _ 1 _ 3 _ 3 _ 3 _ 7 _ 0 1 4 _ 1 will cause 3 entries in nuclide 1, 3 entries in nuclide 3, all entries in nuclide 7, and 1 entry in nuclide 14 to be erased for geometry 2.

Card 1	Column	Description
	1-5	99999

Program needs this card to signal end of erase control cards.

LMERGE Control Cards (None required)

OPERATING INSTRUCTIONS FOR CHSUM

General

The CHSUM routine is used to determine the fractional contributions from each photopeak channel grouping into every other photopeak channel grouping. It accepts the master tape (Data Set 16) prepared by routine BASIC and prepares a tape (Data Set 20) of fractional contributions, decay constants, and conversion factors for use in PROFRM. The output is under switch control.

CHSUM Data Sets

Number	Device	Subroutine	Purpose
16	9-Trk	CHSUM	Output from BASIC containing sorted basic calibration data Input to CHSUM
20	9-Trk	CHSUM	Output from CHSUM containing scatter fractions by geometry and nuclide Input to PROFRM

CHSUM Control Cards

Decay Constants

Card 1	Column	Description
	1-2	L - Number of nuclides for which decay constants are required

Example

_ 7 would indicate that decay constants for 7 nuclides are required.

Card(s) 2 - (L+1)	Column	Description
	1-2	Nuclide number
	4-12	Decay constant associated with nuclide in 1-2

Example

_ 1 _ 0 . 6 9 3 E + 0 1 would indicate that nuclide 1 has a decay constant of 6.93.

Conversion Factors

Card 1	Column	Description
	1-3	Geometry number
	4-6	Group selection number
	7-9	NN - Number of conversion factors for this geometry and group selection

Example

_ _ 2 _ _ 1 _ 3 0 would indicate that for geometry 2, group 1, there are 30 conversion factors.

Card(s) $2 - \left(2 + \left[\frac{NN-1}{6}\right]\right)$	Column	Description
	1-2	Nuclide number
	4-12	Conversion factor
	13-14	Nuclide number
	16-24	Conversion factor
	⋮	

Example

_ 2 _ 1 . 2 8 6 E + 0 1 _ 7 _ 2 . 7 3 6 E - 0 1 would indicate that nuclides 2 and 7 have conversion factors of 12.86 and 0.2736, respectively.

This continues for NN conversion factors with 6 factors/card. Repeat from card 1 for each group selection-geometry combination.

Card 1	Column	Description
	1-5	99999 - Terminates reading of conversion factors

Group Definitions

Card 1	Column	Description
	1-2	Number of group definitions Max 10 (usually 1)

Example

_ 1 would indicate that there is only 1 group defined.

Card 2	Column	Description
	1-2	M - Number of nuclides for this group definition

Example

3 0 would indicate that there are 30 nuclides in this group definition.

Card(s) 3 - (M+2)	Column	Description
	1-4	Beginning channel number
	5-8	Ending channel number
	9-12	Nuclide number

Example

_ 1 2 9 _ 1 3 7 _ _ 1 8 would indicate that channels 129 through 137 inclusive are to be summed for the photopeak area of nuclide 18 for this group definition.

Repeat from card 2 for each group definition.

Group Selections

Card 1	Column	Description
	1-2	Geometry number
	3-4	Number of group selections desired
	5-6	Group selection number 1
	7-8	Group selection number 2
	⋮	

Example

_ 3 _ 1 _ 1 would indicate that for geometry 3, only 1 group selection is desired and its identification is 1.

Repeat card 1 for each geometry.

Card 1	Column	Description
	1-5	99999 - End group selection cards

OPERATING INSTRUCTIONS FOR PROFRM

General

PROFRM takes the tape (Data Set 20) containing the scatter fractions for the various geometries and nuclides, which were calculated in CHSUM, and forms sets of nuclides representative of areas of sampling. Matrices of the scatter fractions for each set of nuclides are inverted and placed on tape (Data Set 10) for use by CALC.

These sets of nuclides are called "nuclide programs" and are defined uniquely by a 5-digit code as follows:

Digit	Purpose
1	A sequence number for programs with this geometry and number of nuclides
2-3	Number of nuclides in this program
4-5	Geometry for this program

PROFRM Data Sets

Number	Device	Subroutine	Purpose
20	9-Trk	PROFRM	Output from CHSUM containing scatter fractions Input to PROFRM
10	9-Trk	PROFRM	Output from PROFRM containing inverted scatter fraction matrices, conversion factors, and decay constants Input to CALC

PROFRM Control Cards

Nuclide Names

Card 1 Column	Description
1-2	MM - Number of nuclides in the system

Example

2 5 would indicate 25 nuclides in the system.

Card(s) $2 - \left(2 + \left[\frac{MM-1}{12}\right]\right)$	Column	Description
		Maximum of 12 nuclides/card; i.e., 25 nuclides would require 3 cards
	1-6	Nuclide name 1 (left-justified)
	7-12	Nuclide name 2 (left-justified)
	⋮	

Example

R U - 1 0 6 Z R N B 9 5 would give names for nuclides 1 and 2.

Program-Unit Combinations

Card 1	Column	Description
	1-2	NK - Number of program-unit combinations

Example

1 3 would indicate 13 program-unit combinations.

Card(s) 2 - (NK+1)	Column	Description
	1-5	Program number
	6-11	Units (right-justified)

Example

1 1 0 0 3 _ _ _ C / G would indicate that program 11003 has units C/G.

Program Definitions

Card 1	Column	Description
	1-5	Program number
	6-9	Group selection number
	10-13	0 - No multiplying constant for conversion factor
		1 - Multiplying constant for conversion factor
	14-22	Multiplying constant if 10-13 has a 1

Example

1 0 3 0 3 _ _ _ 1 _ _ _ 0 would indicate that program 10303 uses group selection 1 and uses input conversion factor.

Card 2	Column	Description
	1-2	Nuclide number of first nuclide desired
	3-4	Nuclide number of second nuclide desired
	⋮	

Example

_ 1 _ 3 _ 7 would indicate that nuclides 1, 3, and 7 are to be incorporated into program 10303.

This continues for as many nuclides as indicated on previous card. Repeat cards 1 and 2 for as many programs as desired.

Card 1	Column	Description
	1-5	99999 - last card of PROFRM

OPERATING INSTRUCTIONS FOR CALC

General

The subroutine, CALC, calculates the intensities of the nuclides present in a sample by the method of simultaneous equations, as well as the error associated with each intensity.

Data Sets

Number	Device	Subroutine	Purpose
4	7-Trk	CALC	Sample spectra prepared on IBM 1401
10	9-Trk	CALC	Output from PROFRM with inverted scatter fraction matrices

No control cards are needed.

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