MASTER

DATA ANALYSIS TECHNIQUES
FOR
GAMMA-RAY SCINTILLATION SPECTROMETRY

R. L. Heath

PHILLIPS
PETROLEUM
COMPANY

ATOMIC ENERGY DIVISION

NATIONAL REACTOR TESTING STATION
US ATOMIC ENERGY COMMISSION

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

PRICE \$.50

Available from the
Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Printed in USA

IDO-16784
AEC Research and Development Report
Physics [REDACTED]
TID-4500 (17th Ed.)
Issued: May 29, 1962

DATA ANALYSIS TECHNIQUES FOR GAMMA-RAY SCINTILLATION SPECTROMETRY

by

R. L. Heath

PHILLIPS
PETROLEUM
COMPANY

Atomic Energy Division
Contract AT(10-1)-205
Idaho Operations Office
U. S. ATOMIC ENERGY COMMISSION

DATA ANALYSIS TECHNIQUES FOR GAMMA-RAY SCINTILLATION SPECTROMETRY

ABSTRACT

The most important area of development in gamma-ray scintillation spectrometry at the present time is the adaptation of digital computer techniques to the analysis of pulse-height spectra. The use of multi-dimensional pulse analyzers capable of storing 256 spectra, each containing up to 256 channels of digital information, has greatly increased the need for automated data processing techniques. As a first step, logical adders and tape input-output systems have been added to modern analyzers to permit some reduction of data within the machine itself. The development of analytical techniques adaptable to machine programming, however, appears to represent the only reasonable solution of the problem. A series of computer programs are described which have been successfully applied to the reduction of pulse-height data using digital computers. The calculations carried out by these programs include: determination of pulse height vs energy response of an NaI detector; calculation of coincidence sum spectra; and the analysis of complex pulse-height spectra to obtain energies and relative intensities of individual gamma rays or relative abundances from a mixture of radionuclides. Measurement problems and instrumental requirements are discussed in detail.

DATA ANALYSIS TECHNIQUES FOR GAMMA-RAY SCINTILLATION SPECTROMETRY

CONTENTS

ABSTRACT	ii
INTRODUCTION	1
DETECTOR RESPONSE PROGRAM	1
COINCIDENCE SUM EFFECTS	5
COMPUTER PROGRAMS FOR ANALYSIS OF COMPLEX SPECTRA	7
PEAK-FITTING PROGRAM	8
GAIN-SHIFT PROGRAM	8
LEAST-SQUARES FITTING PROGRAM	8
APPENDIX I	
Pulse Height vs Energy Response of NaI	10
APPENDIX II	
Multi-Channel Pulse-Height Analyzer Performance Specifications	10
REFERENCES	11

FIGURES

1. Measured pulse-height distributions for single gamma rays used for the determination of the response function of an NaI detector	2
2. Example of the pulse-height spectrum of a single gamma ray showing the effects of scattering and random summation spectrum	2
3. Three-dimensional model of the response of a 3" x 3" NaI(Tl) detector to monoenergetic gamma radiation	3
4. Plot of b_0 , related to photopeak width, as a function of gamma-ray energy	4
5. Detector response to a single gamma ray showing details of structure and method used in computing Compton distribution	5
6. Comparison of computed response to a 1.78-Mev gamma ray with experimental pulse-height distribution	6
7. Pulse-height spectrum of NB-94 comparing experimental spectrum with computed coincidence sum spectrum	6
8. Pulse-height spectrum of gamma rays emitted in the decay of 42-day Pm-148 comparing experimental spectrum with computed spectrum shape	7
9. Example of a normalized least-squares analysis of a five-component spectrum containing 10 gamma rays	9
10. Pulse height <u>vs</u> energy response for a 3" x 3" NaI detector	10

TABLES

I. Measured Values of dL/dE as a Function of Gamma-Ray Energy	10
---	----

DATA ANALYSIS TECHNIQUES FOR GAMMA-RAY SCINTILLATION SPECTROMETRY

R. L. Heath

Introduction

The analysis of pulse-height spectra, using both scintillation and solid-state charged-particle detectors is being applied to a variety of measurement problems in nuclear physics. It should be emphasized, however, that at present the widest application is in the field of gamma-ray scintillation spectrometry. It has been conservatively estimated that over 500 gamma-ray spectrometers are in operation in this country, representing an investment of over 20 million dollars in pulse analyzers, detectors, and associated equipment. This degree of application has been achieved in spite of the difficulties associated with the analysis of the data obtained using these devices. If suitable methods can be devised to permit the use of digital computers for data analysis, applications of the scintillation method will be greatly simplified.

The problem of data analysis, simply stated, is to find a satisfactory method for developing a mathematical transformation between a measured pulse-height distribution and the true energy spectrum emitted by the radiating source. The solution, however, is not simple because there are many complex and variable factors involved in the measurement of pulse-height distributions.

Detailed discussions of this problem have been presented by many authors¹⁻⁴; and many methods⁵⁻¹⁰ have been developed which produce successful results in limited applications. It is evident that none of these techniques represents a solution to all measurement problems. It should be emphasized that pulse-height measurements must be divided into two distinct categories:

(1) continuous gamma-ray spectra; and (2) discrete spectra containing structure which can be associated with individual gamma rays. The latter type, fortunately, is the one usually encountered. In this discussion we will confine ourselves to a consideration of discrete spectra only. The major problems in this area are the measurement of individual gamma-ray energies and intensities and the identification and measurement of the abundance of radionuclides present in a sample. Although the methods to be described were developed primarily for these latter problems, they may also be applied to the analysis of continuous spectra.

A multi-channel pulse-height analyzer presents the pulse-height spectrum as a histogram, each channel representing the number of pulses per unit time whose amplitudes fall into the interval $V + \Delta V$. When the detector is exposed to a radioactive source emitting only gamma rays of one energy, the observed pulse-height distribution is

generally spread over many channels. The relative number of pulses in a given channel represents the probability that a gamma ray from this source will produce a pulse of the corresponding height. The fact that the detection of monoenergetic gamma radiation gives rise to a continuous distribution of pulse heights arises from the fundamental nature of the detection process itself and greatly complicates the problem of analysis. Since counts will be distributed over a large number of channels for each discrete gamma-ray energy detected, if gamma rays of several different energies are present in the spectrum, each channel may contain a contribution from several gamma rays. For this reason, any method with general application to the problem of analyzing experimental pulse-height distributions requires a precise determination of the pulse height vs energy response function for the particular detector and experimental environment used in the measurement.

Detector Response Program

For this reason, considerable effort has been directed toward obtaining a detailed knowledge of the pulse-height response to monoenergetic gamma radiation. To obtain a one-to-one correspondence between the true gamma-ray spectrum from a source and the observed pulse-height distribution, two methods have been employed. One approach has been an attempt to calculate the pulse-height distribution from a knowledge of the basic interactions occurring in the detector. This approach has resulted in the development of very sophisticated Monte-Carlo computer programs as described by Zerby¹¹. The other, or empirical, approach has been to measure the response of a detector to a series of radioactive sources each emitting a monoenergetic gamma ray. Graphical interpolation, between these measured shapes, is then used to obtain the response to any gamma ray. These empirical techniques have been described in the Gamma-Ray Spectrum Catalogue¹² and by P. R. Bell¹³. Fig. 1 shows a typical set of measured pulse-height distributions for single gamma rays. These data have been normalized to the same emission rate using measured photo-fractions.

As pointed out by Zerby¹¹, definite discrepancies exist between the computed pulse-height distributions for single gamma rays and those measured experimentally. While some of these differences may be ascribed to scattering from surrounding material and bremsstrahlung produced in the slowing down of betas emitted by the sources, we do not feel that this can account entirely for the observed discrepancies. It should be stressed that any scheme for analyzing spectrometer data must account for the pulse-height

Fig. 1 Measured pulse-height distributions for single gamma rays used for the determination of the response function of an NaI detector.

distribution actually observed in a given experiment. Either the effects of scattering must be handled independently for each particular geometry or the geometry must be fixed and the scattering component included in the analysis.

As an example of the complex response of a scintillation detector to a single gamma ray, Fig. 2 shows the pulse-height distribution from a 478-keV gamma ray, measured using a 3" x 3" NaI detector. To illustrate the problems to be considered in the analysis of pulse-height spectra, the contributions of Compton scattering from material surrounding the source and detector and from absorbing material which frequently must be inserted to exclude beta radiation emitted by the source are shown. The analysis is further complicated by the fact that the magnitude and shape of the scatter components will depend upon the particular measurement geometry employed. In addition to the spectrum of pulses resulting from the detection of single gamma rays in the detector, we must consider the effect of random coincidence summation of pulses which result from simultaneous detection of two or more gamma rays within the resolving time of the electronic system. This results in a distribution of pulses from zero amplitude to twice the amplitude of the full-energy peak. The probability that a pulse will appear in the random coincidence sum spectrum is proportional to the resolving time of the system and to the input counting rate. The shape and magnitude of this spectrum may be calculated from a convolution of the single spectrum with itself. Details of the method used to compute such spectra are given in a later section.

The relative magnitude of all effects which contribute to the observed pulse-height distribution for a single gamma ray are highly dependent

Fig. 2 Example of the pulse-height spectrum of a single gamma ray showing the effects of scattering and random summation spectrum.

on the experimental arrangement used in a given measurement. For this reason, our feeling is that the best approach in the development of data processing techniques for a large majority of users requires standardization of source-detector geometry. These complicating effects are then included as a part of the standard response function.

To facilitate the application of sophisticated analytical techniques to the analysis of pulse-height spectra, computer techniques have been developed for the calculation of the pulse-height response of an NaI scintillation detector as a function of gamma-ray energy. This method of calculating a response function may be applied to any experimental arrangement and detector.

The approach used is a systematic extension of the graphical interpolation scheme which has been used by nuclear spectroscopists for some

time. If we consider that a sufficient number of radioactive sources may be obtained which decay with the emission of a single gamma ray, then the pulse-height spectrum for a gamma ray of any energy may be determined by a suitable interpolation scheme. First, a number of carefully measured spectra are obtained for single gamma rays as shown in Fig. 1. Using these pulse-height distributions, we may, by interpolation, construct a three-dimensional model of the response of an

NaI scintillation detector to monoenergetic gamma rays. Fig. 3 illustrates such a response function for a 3" x 3" NaI detector. Pulse-height distributions for single gamma rays, normalized to the same emission rate, are arranged with gamma-ray energy as the third coordinate. A plane perpendicular to the energy axis through a given point will intersect this surface to form a curve. This curve represents the detector response to a gamma ray of that energy. The generation of such a

Fig. 3 Three-dimensional model of the response of a 3" x 3" NaI(Tl) detector to monoenergetic gamma radiation.

surface by means suitable for machine programming requires an analytical representation of the experimentally determined pulse-height spectra. This is accomplished by representing a pulse-height distribution as a sum of several analytic functions. If we examine the detailed structure of pulse-height distributions obtained from mono-energetic gamma rays, as shown in Fig. 3, it is apparent that there are several features which vary in a uniform manner with gamma-ray energy. All features of the spectra can be interpreted from a knowledge of the three basic processes by which a photon interacts with the detector; i.e., photo-electric, Compton, and pair interactions. The most prominent feature, the photopeak, is essentially gaussian in shape with some asymmetry on the low-energy side due to multiple processes occurring in the detector and small-angle Compton scattering from absorbing materials frequently used to prevent detection of beta radiation emitted by the source. This peak is represented by a gaussian of the form

$$y = y_0 e^{-\frac{(x-x_0)^2}{b_0}}$$

where y_0 is the maximum amplitude of the peak, x_0 is the midpoint of the distribution on a pulse-height scale, and b_0 is related to the full width of the gaussian shape at half-maximum (resolution). The convention adopted is to fit the experimental photopeaks on the high-energy side and to determine the parameters of the gaussian by least-squares techniques. The observed deviation from a true gaussian shape on the low-energy side is treated as though it belonged to the Compton distribution. The experimentally determined photopeaks are fitted in this manner and the resultant values for the width parameter, b_0 , are fitted to a polynomial in pulse height (energy). The results of such a calculation for a 3" x 3" NaI detector are shown in Fig. 4. From a knowledge of b_0 and

Fig. 4 Plot of b_0 , related to photopeak width, as a function of gamma-ray energy.

the normalized area of the gaussian as a function of pulse height (energy) it is therefore possible to calculate the shape and magnitude of the photopeak for a gamma ray of any energy. It should be noted that the pulse height vs energy scale must include the effect of non-linearity in the energy response of NaI. This effect is discussed in Appendix I.

The remaining portion of the pulse-height distribution is due largely to the Compton process which produces a distribution of pulses from zero amplitude up to a sharp cut-off representing maximum energy transfer between the photon and a recoil electron. This point on the energy spectrum is termed the Compton edge (E_C) and is given by the following relationships:

$$E_C = E_\gamma - \frac{E_\gamma}{1 + \frac{2 E_\gamma}{mc^2}}$$

Other prominent features of the spectrum are the back-scatter peak which results mostly from photons Compton scattered through $\sim 180^\circ$ from material surrounding the detector, the two pair peaks resulting from the escape of one or both annihilation quanta, and the 0.511-Mev peak from annihilation radiation. The position of these peaks on the pulse-height (energy) scale is given by the following relationships:

$$\text{back-scatter peak } E_b = \frac{E_\gamma}{1 + \frac{2 E_\gamma}{.511}}$$

$$\text{single-escape peak } E_{p1} = E_\gamma - .511 \text{ Mev}$$

$$\text{double-escape peak } E_{p2} = E_\gamma - 1.02 \text{ Mev}$$

Fig. 5, which shows the pulse-height distribution for a 1.78-Mev gamma ray, illustrates the details described above. As shown in this figure, the portion of the spectrum below the full-energy peak is divided into 5 segments $L_1 \dots L_5$ which are bounded by $O, X_b, X_{p1}, X_{p2}, X_c$, and X_o . Each segment is then fitted to a series of the form:

$$y = a + bx + \sum_{K=1}^N b_K \sin \frac{K \pi X}{L}$$

where the lower channel limit is taken as the origin and the upper limit, L , is the upper bound of the segment being fitted. In order to ensure a smooth fit at the segment end points, a region of overlap is included in the fitting procedure. The number of terms, k , in the expansion is chosen to give a certain amount of smoothing to the data points. It should be noted that below the pair threshold (1.02 Mev) only three segments with the

Fig. 5 Detector response to a single gamma-ray showing details of structure and method used in computing Compton distribution.

bounds X_b , X_c , and X_0 are required to adequately describe the Compton distribution.

The next step is to determine the count-rate, $[N(E)dE]$, in a given channel, as a function of gamma-ray energy. These points, which represent a section through the 3-dimensional surface perpendicular to the pulse-height axis, are then fitted to a polynomial in gamma-ray energy. The number of terms in this polynomial is adjustable in order to give the most reasonable fit to the data points. Thus a set of polynomials in energy are obtained, one for each channel on the pulse-height scale. This process is repeated for each segment of the Compton distribution. To simplify this interpolation with a limited number of measured spectra, each group of segments is first stretched so that each segment has the same length for all gamma-ray energies. This preserves the fine structure which is varying with gamma-ray energy; i.e., back scatter and pair peaks. Since the number of segments required to describe the Compton distribution varies with E_γ , the program is divided into three sections. These sections treat the following ranges of gamma-ray energy: 0.020-0.300, 0.300-1.114, and 1.114-4.00 Mev.

To obtain the pulse-height distribution for a given gamma ray, channel count rate values are computed from the set of polynomials representing

$N(E)dE$ vs E for each channel. These values for each segment are combined with a photopeak calculated from values of b_0 and A (area of the gaussian) obtained from polynomial fits of these quantities as a function of E_γ .

Fig. 6 shows a comparison between an experimental pulse-height distribution for a single gamma ray and the calculated shape for a gamma ray of the same energy obtained from the shape generation program. The two spectra are displaced on the vertical scale for comparison. It is seen that excellent agreement is obtained. All details of the spectrum are accurately produced. Agreement of all points on the two spectra are within the range of expected statistical variations in the experimental data.

With this precision it is possible to calculate the pulse-height distribution expected from any photon for a given set of experimental parameters.

Coincidence Sum Effects

An additional complication in the analysis of complex gamma-ray spectra occurs when the decay of a radioactive element gives rise to two or more coincident gamma rays. In this case the pulse spectrum observed in a detector will also include

Fig. 6 Comparison of computed response to a 1.78-mev gamma ray with experimental pulse-height distribution.

a distribution of pulses known as the coincidence sum spectrum which results from the simultaneous detection of two coincident gamma rays. This spectrum will contain pulses of all sizes from the minimum detectable height to the sum of the maximum pulse height obtainable from the individual gamma rays. The most prominent feature of this spectrum is the so-called sum peak which results from coincident detection of pulses from the photo-peaks of the two coincident gamma rays. This effect has been well described in the literature ^{3, 4}.

In principle it is possible to calculate both the shape and magnitude of this summation spectrum. Let us consider a two-gamma-ray cascade. Using the program previously described we may compute the detector response to the individual gamma rays for a given detector and experimental environment. If the computed pulse-height distributions for the two coincident gamma rays are normalized to one disintegration of the source, then values of $N(E)dE$ may be determined for each spectrum which represent the probability of detecting the corresponding gamma ray as a count in a given channel of the pulse-height spectrum. Pulse-height spectra for the two coincident gamma rays may be designated as $G_1(X)$ and $G_2(X)$.

The probability of detecting gamma 1 in channel a and gamma 2, from the same disintegration, in channel b is the product $G_1(a)G_2(b)$. This product is then the probability of producing a pulse in the sum spectrum in channel $(a + b)$. The total contribution to any channel, c, of the sum spectrum is then the sum of contributions from all pairs of channels for which $a + b = c$. This may be expressed in the following manner:

$$S(c) = \sum_{n=1}^c G_1(n) \cdot G_2(c-n) \quad (1)$$

A Fortran program has been written which performs this summation for 2-, 3-, or 4-gamma-ray cascades, using as input data, calculated spectra from the previously described spectrum generation program. The output spectra from the generation program are

normalized to unit emission and corrected for the non-linear energy response of NaI.

One further correction is necessary to obtain the correct intensity for the sum spectrum relative to the cascade gamma rays. Each pulse which appears in the sum spectrum represents a count missing from the pulse-height spectra of each of the summing gamma rays.

As an example of the results of this program, Fig. 7 shows the experimental spectrum of two coincident gamma rays together with the computed coincidence sum spectrum. Both the shape and intensity are reproduced with considerable precision. With the addition of this program we now have the capability of computing the pulse-height distribution representing the gamma-ray spectrum emitted by any radioactive nuclide. This requires only a knowledge of the decay scheme and may be applied in principle to any detector or experimental arrangement.

Fig. 7 Pulse-height spectrum of Nb-94 comparing experimental spectrum with computed coincidence sum spectrum.

Fig. 8 shows a comparison between the experimental pulse-height spectrum of 42-day Pm¹⁴⁸ and the computed spectrum obtained using the programs described above. The computed spectrum was generated from data on the decay scheme of this nuclide obtained at this laboratory. This is a complicated case involving 17 gamma rays with many double and triple cascades. All features of

Fig. 8 Pulse-height spectrum of gamma rays emitted in the decay of 42-day Pm-148 comparing experimental spectrum with computed spectrum shape (calculated spectrum intensity displaced arbitrarily for clarity).

both the singles and coincidence sum spectra are reproduced with high precision. The two spectra have been displaced on the vertical axis for comparison. With the exception of the two low-energy peaks, which were not included in the calculation, all points on the computed spectrum agree quite well with the experimental data. It is felt that this is adequate demonstration that the response function generation program will yield sufficient precision to apply it to more complex analytical techniques for analyzing pulse-height data.

Computer Programs for Analysis of Complex Spectra

Using the two programs previously described it is possible to compute quite accurately the

pulse-height spectrum for a single gamma ray or the entire pulse-height spectrum to be expected from the decay of any nuclide. In order to apply the results obtained from these programs to the analysis of complex spectra, it is necessary to develop a method which will take into consideration the many problems associated with experimental measurements of pulse-height spectra. The principal items to be considered are:

- (1) The data obtained from radioactive decay are statistical in nature.
- (2) Gain and zero shift in the spectrometer or detector must be considered in order to establish a proper pulse-height-energy relationship.

(3) The treatment of errors in the data must be included to provide information on the validity of the result.

A method of analysis has been developed for application to a variety of measurement problems. This method was designed to be applied to the analysis of spectra which contain a number of components with some information available to identify the major features observed in the spectrum. For the purpose of this discussion we may consider that the method may be divided into three separate operations:

(1) A composite pulse-height spectrum is examined to determine the number of photopeaks present in the sample and to locate them accurately on the pulse-height scale.

(2) Using a well resolved photopeak, present in the sample, corresponding to a gamma ray of known energy, the energy-pulse-height scale is determined and the entire spectrum is then normalized to a standard scale to eliminate the result of gain shift in the detector or analyzer. The standard reference line may either be a well known gamma ray emitted by one of the constituents in the source or a standard which is measured simultaneously with the spectrum to be analyzed.

(3) Energy values for the major peaks, obtained from the first two operations, serve as an initial guess as to the composition of the sample. Spectra of individual gamma rays or nuclides are then obtained from the detector response program or from standard libraries of response functions to radioactive nuclides. With all spectra properly normalized to the same pulse-height scale, a least-squares fitting technique is applied to determine the relative magnitudes of each constituent in the sample.

Peak-Fitting Program

This program permits any number of channels in a given pulse-height spectrum to be fitted by the method of least squares with a combination of gaussian curves, straight lines, and/or exponentials. Thus a region in a given pulse-height distribution that has been identified as a single or composite photopeak can be fitted with one or more gaussian shapes where any or all variables of the gaussians (b_0 , y_0 , x_0) can be fixed or allowed to vary. A combination of one or more additional functions may be added to account for contributions in this region from higher energy gammas. This program is written in Fortran and can successfully handle up to 8 different functions in one fit. This procedure can locate the position of a typical photopeak to within ± 0.02 channel. This corresponds to 0.2 kev for a 10-kev/channel scale.

Gain-Shift Program

This program is used to correct for shifting in the gain or zero of the spectrometer. To achieve this shift on the pulse-height scale, the

pulse-height distribution is represented analytically by a series of polynomial least-squares fits to sets of 3 channels. The result of these successive fits around the original points is to provide interpolated values of count rate. Conserving the total area under the distribution, the original pulse-height scale is multiplied by the appropriate factor to place the photopeak at the proper position on the standard pulse height vs energy scale.

Values of channel count rate are then calculated for the new channel positions by interpolation from the set of computed polynomials. The only modification of the original data, for small scale shifts, is to produce a slight smoothing of the data.

Least-Squares Fitting Program

The basic problem in the analysis of complex pulse-height spectra, as previously outlined, is to determine the contribution to a given channel, i , from each component in the spectrum. Consider a pulse-height spectrum consisting of i channels numbered $a-i-n$, containing j components, (either gamma rays or complete spectra of nuclides) numbered $1-j-c$. Further, consider that standard spectra measured under identical conditions with the composite are available for comparison. Then the count rate in channel i from the j th component will be S_{ij} and the total count rate in channel i from the composite of all components will be C_i . If the prerecorded standards are all normalized to a given disintegration rate from the source, then we must include a normalizing factor α_j , which is the ratio of the intensity of component j in the composite to the normalized standards. We may then write:

$$C_i = \sum_{j=1}^c \alpha_j S_{ij} + R_i$$

where R represents the random error.

In the sense of least squares, the sum of the squares of the residuals R_i are minimized over all channels.

$$\text{minimize } \sum_{i=a}^n (C_i - \sum_{j=1}^c \alpha_j S_{ij})^2 w_i$$

where w_i is the weighting factor.

The resultant set of equations are solved to obtain values for the α_j 's using matrix techniques. The program thus uses all available information from each spectrum to obtain the best fit over all channels. Standard deviations may be calculated and errors assigned to the result. A weighted fit of this type can be used to analyze very poor data with considerable accuracy.

As an example of the application of these techniques, Fig. 9 shows the results from the analysis of a complex spectrum containing five components and a total of 10 gamma rays. In order to demonstrate the precision of the method, the 5 standards were measured individually and then

Fig. 9 Example of a normalized least-squares analysis of a five-component spectrum containing 10 gamma rays.

measured together to form the composite. The calculated values for the intensity of each component are tabulated on the figure and compared with intensities obtained from the five components measured separately. Deviations are less than 1% in all cases, even though the composite spectrum is quite complex. The residuals resulting from the least-squares analysis, in units of R/σ , are shown, plotted for each channel, at the bottom of the figure. The maximum deviation for any point is seen to be less than 2σ .

This method can be applied to the determination of gamma-ray energies and intensities for

nuclear research applications and to the quantitative analysis of mixtures of radionuclides. In our own laboratory it is being used for routine analyses of radiochemical samples and unknown mixtures. It is currently being adapted to the reduction of data obtained from a multi-dimensional pulse-height analyzer. The response function program can easily be adapted to large matrix solutions applicable to the analysis of continuous gamma-ray spectra.

Since the performance requirements for equipment to be applied to this type of analysis are somewhat important, a list of the minimum acceptable specifications are given in Appendix II.

Appendix I

Pulse Height vs Energy Response of NaI

The non-linear energy response of NaI to gamma radiation has been discussed in detail by several authors ¹⁴⁻¹⁷. In any systematic treatment of data from NaI spectrometers it is essential that an absolute pulse height vs energy scale be established. For a given experimental arrangement this scale depends on the light output from the detector as a function of E_γ , (dL/dE) ; and the linearity of the electronic system. For this reason a determination was made of the pulse height vs energy scale for the experimental arrangement employed in the studies described above. In order to provide an absolute pulse-height scale for data obtained on a multi-channel pulse-height analyzer the finite channel width must be taken into account. Since the observed number of counts in a given channel represents an integral from E to $E + \Delta E$, the average pulse amplitude is most nearly represented by the midpoint of the channel. Thus, for example, channel 100 must span from 99.5 to 100.5 pulse-height units. This requires that the zero on the multi-channel analyzer be established on this scale as -0.50 channels.

A series of radioactive sources emitting gamma rays of precisely known energies was measured together with the Cs^{137} gamma ray as a reference. The absolute pulse-height scale described above was used for these measurements. Least-squares fitting techniques were used to determine the photopeak positions to an accuracy of ± 0.03 channels. The zero position and integral linearity of the electronic system were determined with a calibrated pulser. For the system used (A-8 amplifier and ND 130A analyzer), deviations from a straight line passing through the origin (channel zero) did not exceed ± 0.05 channels over a range of channel 5 to channel 250. The results of these measurements are tabulated in Table I. Fig. 10 shows a

Table I

Measured values of dL/dE as a function of gamma-ray energy

	Gamma-Ray Energy	PH/ ΔE
Am^{241}	0.05957	1.158
La^{177}	0.11297	1.0943
	0.20836	1.0435
I^{131}	0.36447	1.019
Be	0.478	1.011
Bi^{207}	0.5695	1.006
Cs^{137}	0.66162	1.000
Y^{88}	0.8989	0.9893
Bi^{207}	1.0637	0.9854
Zn^{65}	1.114	0.9828
Y^{88}	1.837	0.9677

Fig. 10 Pulse height vs energy response for a 3" x 3" NaI detector.

comparison of the experimental values obtained for dL/dE vs E_γ with those of Engelkemeir ⁵ and of Managan ⁴. Results for the detector used agree very closely with those of Engelkemeir. Similar measurements, using several commercial detectors, have indicated some variation in response below 100 kev. For this reason, the determination of the pulse height vs energy response for a given detector must include a measurement of the non-linear response for that detector.

Appendix II

Multi-Channel Pulse-Height Analyzer Performance Specifications

The specifications listed below represent a current estimate of minimum performance requirements for equipment to be applied to precision data reduction using computer techniques.

(1) Memory Capacity

To permit data reduction an analyzer should have a minimum of 400 channels, with 200 or 256 channels to be used for a normal spectrum. The remaining group to be applied to data reduction.

(2) Integral Linearity of ADC

To be equal to or better than ± 0.1 channel, when defined as maximum deviation in any channel from a straight-line plot of channel position vs input pulse amplitude.

(3) Differential Linearity

Defined as deviation in window width. To be better than $\pm 1\%$ from channel 3 up, as measured with a sliding-ramp pulser.

(4) Noise Modulation

Expressed as noise level of pulse amplifier in percent of full output. To be less than

the equivalent of 1/2 channel for a normal gain setting (25% of full gain).

(5) Accuracy of ADC

ADC should be able to store >98% of pulses in one channel, over 1/3 of the width of one channel, using a noise-free relay pulser and an amplifier with the noise characteristics described above.

(6) Count-rate Shift

Less than 0.05 channels from 0 to 20,000 input pulses/sec (as measured with pulser and source simultaneously) using a double-differentiated amplifier such as the type A-8.

(7) Accuracy of Live Timer

The accuracy of the live-timer should be better than $\pm 0.2\%$ for counting periods of 1 second or greater. This implies electronic gating, pulse synchronization, and crystal controlled time-oscillator.

(8) Zero Stability

To be better than ± 0.1 channels/day for a temperature range of $\pm 5^\circ\text{F}$.

References

1. N. H. Lazar, "Analysis of gamma-ray scintillation spectra for quantitative photon intensities", Proc. Sixth Scintillation Counter Symposium, IRE Trans. on Nuclear Science, Vol. NS-5 pp. 138-146; December, 1958.
2. K. Linden and N. Starfelt, "Scintillation spectroscopy of continuous gamma and X-ray spectra below 1 Mev", Arkiv Fysik, Vol. 7, pp. 427-457; 1954.
3. W. R. Dixon and J. H. Aitken, "The resolution correction in the scintillation spectrometry of continuous X-rays", Canad. J. Phys., Vol. 36, pp. 1624-1633; December, 1958.
4. W. R. Barrus, "Unscrambling scintillation spectrometer data", Proc. Seventh Scintillation Counter Symposium, IRE Trans. on Nuclear Science, Vol. NS-7, pp. 102-111; June, 1960.
5. H. I. West and B. Johnson, "Scintillation counter γ -spectra unfolding code for the IBM-650 computer, "Proc. Seventh Scintillation Counter Symposium, IRE Trans. on Nuclear Science, Vol. NS-7, pp. 111-115; June, 1960.
6. J. H. Hubbell and N. E. Scofield, "Unscrambling of gamma-ray spectrometer pulse-height distributions", IRE Trans. on Nuclear Science, Vol. NS-5, pp. 156-158; December, 1958.
7. J. F. Mollenauer, "A computer analysis for complex sodium iodide gamma spectra", University of California report No. UNRL-9748; August, 1961.
8. L. E. Fite, D. Gibbons, and R. E. Wainerdi, "Computer coupled automatic activation analysis", Activation Analysis Laboratory, A & M College of Texas, Annual Report (TEES-2671-1); May, 1961.
9. H. Childers, "Analysis of single crystal pulse-height distributions", Rev. Sci. Instr., Vol. 30, No. 9, pp. 810-814; September, 1959.
10. R. L. Heath, "Data processing techniques for routine application of gamma-ray scintillation spectrometry", Proceedings of the Total Absorption Gamma Ray Spectrometry Symposium, U. S. AEC Report TID-7594; May, 1960.
11. C. D. Zerby and H. S. Moran, "Calculation of the pulse-height response of NaI scintillation counters", U. S. AEC Report ORNL-3169; 1962 (unpublished).
12. R. L. Heath, Scintillation spectrometry gamma-ray spectrum catalogue, U. S. AEC Report IDO-16408; (1958).
13. P. R. Bell, "The scintillation method", Beta- and Gamma-Ray Spectroscopy, edited by K. Seigbahn, Interscience Publishers, Inc., New York; (1955).
14. W. Managan and C. E. Crouthamel, "Extrinsic variable", Applied Gamma-Ray Spectrometry, edited by C. E. Crouthamel, Pergamon Press, New York; (1960).
15. D. Engelkemeir, Rev. Sci. Instr., Vol. 27, No. 8, pp. 589; (1956).
16. C. D. Zerby, A. Meyer, and R. B. Murray, "Intrinsic line broadening in NaI(Tl) gamma-ray detectors", Nucl. Instr. and Methods, 12, pp. 115; (1961).
17. P. Iredale, "The non-proportional response of NaI(Tl) crystals to γ -rays", Nucl. Instr. and Methods, II, pp. 336; (1961).