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PUERTO RICO NUCLEAR CENTER

COMPUTER AIDED DECOMPOSITION OF GAMMA SPECTRA EMITTED BY CERTAIN RADIOACTIVE NUCLIDES

By

Aviva E. Gileadi and Nallagounder Kuppusamy

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University of Puerto Rico
Mayaguez Campus

COMPUTER AIDED DECOMPOSITION OF GAMMA SPECTRA EMITTED BY MIXTURES OF CERTAIN RADIOACTIVE NUCLIDES

by

Nallagounder Kuppusamy

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ABSTRACT

A computer code PRGA, written in FORTRAN IV, has been developed in order to perform decomposition of certain gamma spectra obtained by means of Ge(Li) diode equipped multichannel data acquisition system. The code uses spectra of the individual standards and those of the mixtures as input data and calculates the mixing factors pertinent to each component, using linear least squares fit techniques.

PRGA is presently operative on both the IBM-360/40 and the PDP-DEC-10 computers at the Computer Center of the University of Puerto Rico, Mayaguez Campus. The code includes corrections designed to account for deadtime losses, natural and spectral background, decay and counting statistics. Using source mixtures with premeasured mixing factors, specially prepared for the purpose of comparisons, it was found that the premeasured and PRGA computed mixing factors are in satisfactory agreement.

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CHAPTER I

INTRODUCTION

During the last decade or so the development of lithium drifted germanium detectors requiring cryogenic techniques and associated low noise electronics and the use of such detectors with various sophisticated data acquisition systems, featuring such items as multichannel analysers with large memories, often interfaced with digital data processors - made a considerable impact on experimental photon spectrometry.

Improvement of the experimental techniques resulted not only in higher accuracy, and better resolution, but also in an unprecedentedly large quantity of available gamma spectral data. In order to match the efficiency of data acquisition to that of converting these data into meaningful information: numerical techniques are required.

Such techniques not only provide considerable savings in time but also significantly improve the accuracy of data analysis.

Ideally even the most complex spectra should be analysable , qualitatively as well as quantitatively, with little or no man-machine interference. However, before such rather ambitious objective could be achieved, several simpler partial programs need to be developed and checked out, each of them designed to perform a partial task in the framework of complete analysis. Once all the partial programs are operative, they are integrated into a complete system, capable to determine background, to resolve and fit complex peakgroupings, to determine the energies and intensities of the gamma rays as well as the amount of source nuclides within the analysed mixture.

The Nuclear Engineering Department of the University of Puerto Rico, recently acquired a high resolution gamma detecting system consisting of a lithium drifted germanium detector, (manufactured by Princeton Gamma-Tech) to be used in conjunction with a multichannel analyser (model No. 88-0415 manufactured by Nuclear Data, Inc.). This system is to be used in a rather extensive neutron activation project planned by the Department.

It is felt that a reliable and efficient computer code-written especially for analysing data, acquired by the above described, Ge(Li) diode equipped, gamma spectrometer would be a welcome contribution to the departamental efforts in the field of neutron activation.

With this general objective in mind, and as a first step towards it - in the present thesis a computer code PRGA was developed that is capable to decompose gamma spectra emitted by certain source mixtures

PRGA is written in FORTRAN IV and is operative on the IBM-360/40 Computer at the University of Puerto Rico, Mayaguez Campus Computing Center. The code has been tested on gamma spectra emitted by several different premeasured source mixtures and its performance has been found satisfactory.

All gamma spectra used in testing the PRGA code were acquired by means of the Ge(Li) diode equipped gamma ray detector system of the Radioecology Division of the Puerto Rico Nuclear Center, the description of which is given in Chapter V.

CHAPTER II

REVIEW OF LITERATURE

Gamma ray spectrometry is among the most versatile, most important and most frequently applied methods of modern science, accordingly, the professional literature pertinent to that subject cannot be exhaustively reviewed within the scope of the present thesis. An attempt is made, however, to briefly review a selected set of outstanding original papers on the subject of computerized decomposition techniques ("unscrambling") of gamma spectra.

J. MILLS (1) has developed a computer program AUTSPAN for analysis of complex gamma spectra obtained with Ge(Li) detectors. The program contains options for data smoothing, peak location and determination of peak parameters.

G. D. ATKINSON Jr., J. B. WHITWORTH and S. J. GAGE (2) have written a code, NAA, which seeks out peaks, locates the peak maxima, predicts Compton knees, single and double escape peaks and identifies the unknown nuclides present. The code is written in FORTRAN IV and operative on the CDC-6600 computer.

JEAN KERN (3) after a review of the most accurately known gamma calibration lines, presented a method of obtaining new secondary standards of similar precision, but extending to higher energies. Her method uses simultaneously pair peak method, energy combinations and experimental nonlinearity curves.

W. C. WHITE, M. B. SHAPIRO and A. W. PRATT (4) applied linear programming to decomposing spectra.

J. T. ROUTTI and S. G. PRUSSIN (5) have written a Fortran program which searches and fits the single and multiple peaks. In addition their code performs the energy, efficiency, line shape calibration, and error estimates.

The program given by R. G. HELMER, R. L. HEATH, M. PUTNAM and D. H. GIPSON (6) computed the gamma ray energies and intensities and provided an estimate of uncertainty, by fitting the gamma ray peaks to a Gaussian and applying a correction for the nonlinearity of the electronic system, for the data obtained both by NaI(Tl) and Ge(Li) detector systems.

R. GUNNINK and J. B. NIDAY (7) have constructed an elaborate program, GAMANAL, which determines the background, the energies and intensities of the gamma spectrum and the amounts of the source nuclides present in the mixture.

Taking selected channels of data of pulseheight distribution F. D. KNIGHT (8) applied the method of simultaneous linear equations to find the abundance of the concerned nuclides present with the knowledge of the component spectra and the composite spectrum.

The computer program, GAUSS-V, developed by R. G. HELMER and M. H. PUTNAM (9) locates the gamma ray peaks, determines the peak parameters, finds the gamma ray intensities and energies applying nonlinear least square curve fit for the pulse height distribution obtained with a Ge(Li) detector system.

M. CIAMBI, L. DADDI and V. D. ANGELO (10) have fitted the gamma ray photo peaks to a Gaussian, using a maximum probability method and compared the results with those obtained by the least square method.

W. W. BLACK (11) used correlation techniques to locate the photo peaks, he analysed pulse height distributions obtained with Ge(Li), Si(Li), and NaI(Tl) detectors. He also applied this method to data obtained by means of neutron time of flight analysers.

An early paper of J. H. HUBBELL and N. E. SCOFIELD (12) deals with "unscrambling" experimental gamma spectra using matrix techniques.

WALTER R. BURRUS (13) used Fourier techniques to obtain the "unscrambled" spectrum and has assigned meaningful error-estimates to the results.

H. I. WEST Jr. and B. JOHNSON (14) discussed an unfolding code for the IBM 650. They described a procedure for removing scale and energy dependency.

R. L. HEATH, R. G. HELMER, L. A. SCHMITTROTH and G. A. CAZIER (15) used interpolation between experimental spectra, to calculate pulse height distributions obtained by NaI(Tl) crystal system for sources emitting only one gamma ray.

R. J. GEHRKE, J. E. CLINE and R. L. HEATH (16) developed a precise method to determine both the relative detection efficiency and the linearity of the Ge(Li) detector system.

R. G. HELMER, R. G. GREENWOOD and R. J. GEHRKE (17) have measured the energies of gamma ray photons from 24 nuclides using a Ge(Li) detector and have applied correction for the nonlinearity of the detector system and for the recoiling nucleus.

MARIE PUTNAM, D. H. GIPSON, R. G. HELMER and R. L. HEATH (18) developed a computer program which performs a nonlinear least squares fit to pulseheight distributions, obtained both from NaI(Tl) and Ge(Li) detector systems. Their program fits the data to their modified Gaussian functions and prints out the peak parameters.

The program described by R. L. HEATH, R. G. HELMER, L. A. SCHMITTROTH and G. A. CAZIER (19) smoothens the gamma ray spectra, applies correction to the gainshift of the detector system and fits the data Gaussians using linear least squares criterion.

H. BABA, H. OKASHITA, S. BABA, T. SUZUKI, H. UMEZAWA and H. NATSUME (20) constructed a program to analyse gamma ray spectra obtained by using a Ge(Li) detector. The analysis is based on the first derivative method associated with a number of peak shape tests. The effects of data smoothing and change in peak width were studied.

K. LOW (21) attempted to find the contribution from known nuclides in a complex gamma ray spectrum, using least squares method.

W. FILIPPONE and F. J. MUNNO (22) discussed the influence of deadtime losses on the magnitude and shape of the composite spectrum obtained by means of a multichannel analyser. They presented a relation including appropriate corrections.

R. G. WAGGENER, L. F. ROGERS and P. ZANCA (23) Health Physics published a matrix method to correct for non linear energy response of NaI(Tl) spectrometer systems.

R. K. GILLETTE (24) has developed a computer program for the analysis of neutron activation data containing an option for smoothing gamma spectra.

J. I. TROMBKA (25) developed a least squares fitting technique for the analysis of complex gamma ray pulseheight spectra, synthesizing the polyenergetic distribution from a series of monoenergetic components in the incident beam.

H. P. YULE (26) published an exhaustive review paper on computation of experimental results in activation analysis.

D. D. TUNNICLIFF and G. E. A. WYLD (27) have developed a code, based on linear least squares, that computes directly the weights of the components in a source mixture.

CHAPTER III

THEORETICAL BACKGROUND

In the literature numerous methods have been described, for computer aided analysis of gamma ray spectra obtained with Ge(Li) or NaI(Tl) detector-systems. In most of these methods the analysis is performed with the aid of calculated or measured response functions corresponding to monoenergetic photons or individual isotopic components of the sample.

The physical and statistical phenomena determining the response of a semiconductor type detector to a monoenergetic gamma photon, incident on it, are quite complex and the accurate fundamental calculation of peak shapes for the purpose of spectrum analysis is quite difficult. For this reason and because the peak shape may be sensitive to small variations in experimental parameters, it is desirable to determine a mathematical representation of the composite spectra directly from measured data.

In order to assign a suitable functional representation, it is important to examine some of the factors that determine the peak shape. The primary factor determining the width of a photo peak is the statistical fluctuation in the division of the observed energy between ionization and heating of the crystal lattice. This gives rise to a Gaussian distribution with a small specified width. The extent to which this distribution is reflected upon the experimental data, taken with a particular system,is dependent on a number of factors. The material properties and the impurities in the detector affect the charge collection and the electronic noise associated with the leakage current.

The combined effect of incomplete compensation of impurities and incomplete charge collection worsen the resolution and give rise to low energy tailing of the photo peak. At low energies the contribution to the resolution from preamplifier noise is important and at higher energies the inabilities of the amplifier and the pulseheight analyser begin to affect the line width, especially when long counting times are involved. Although the use of the digital gain stabilizer can decrease the broadening, the stabilization may itself change the shape of the peak.

Finally random summing of pulses at high counting rates broadens the peaks and gives rise to tailing. The continuum under the peaks is due to Compton distribution from higher energy gamma rays and general counting background. Without detailed knowledge of the composition of the spectrum we can only say that the continuum in the short interval under one peak or a cluster of peaks is, except for statistical fluctuations, a continuous smoothly varying function of energy. Such a function can be approximated with a polynominal.

Among the several techniques used in computer aided decomposition of complex gamma spectra three major ones will be discussed briefly, following references 6 and 11:

(a) Linear least squares technique

(b) Non linear least squares technique

(c) Correlation technique

In the linear least squares technique the pulse height spectrum due to a polyenergetic distribution of gamma rays is synthesized by using a series of normalized pulse height distributions, due to either monoenergetic components or to the pulse height distribution characteristic of various single nuclides in the source-mixture. Each of these pulse height distributions is weighted so that their sum is a best fit based upon a least squares criterion to the experimentally determined polyenergetic pulse height distribution due to the source mixture under consideration.

It is assumed that the data points in a portion of the experimental spectrum can be represented by a function $\bar{y}_i$ which depends on a set of parameters p_k. The purpose of a least squares fit is to find the value of the parameters, which minimizes the sum of the squares of the deviations of the data from the function; that is

$$R^2 = \sum_i w_i \left[y_i - \bar{y}_i(p_k) \right]^2 \qquad (3.1)$$

is minimized. The $\bar{y}_i$s are experimental data points, the w_i are the weights associated with the y_i, and the $\bar{y}_i(p_k)$ are the values calculated from the function used to represent the data. The summation is overall the data points used in the fit.

A necessary condition for R^2 to be a minimum, as a function of the p_k, is that the set of equations

$$\frac{dR^2}{dp_k} = 0 \qquad \text{for } k = 1,2, \ldots N \qquad (3.2)$$

hold simultaneously for all values of k.

If $\bar{y}_i$ is a linear function of the parameters p_k, the problem is a linear least squares problem, and expressions can be derived for the "best" value of the parameters. However, if the function is not linear in the parameters, no explicit solution of the equations exist. In this case a method of "linearization" may be used.

One such method of linearization is that of Gauss. This method consists of linearizing the function with respect to a set of parameters δp_k by the use of a truncated Taylor series. Initial estimates, p_k^0, of the parameters must be available in order to evaluate the derivatives in the expression; that is:

$$p_k = p_k^0 + \delta p_k \quad \text{and} \quad \bar{y}_i(p_k) = \bar{y}(p_k^0) + \sum_k \frac{\partial \bar{y}_i}{\partial p_k} \delta p_k + \cdots$$

plus expressions containing higher derivatives, where the derivatives are evaluated at the initial values of the parameter.

The function $\bar{y}(p_k)$ is a linear function of the p_k if all derivatives beyond the first are neglected. This gives

$$R^2 = \sum_i w_i \left[y_i - \bar{y}(p_k^0) - \sum_k \frac{\partial y_i}{\partial p_k} \delta p_k \right]^2 \qquad (3.3)$$

The conditions which now determine the minimum are given by

$$dR^2/d(\delta p_k) = 0 \qquad \text{for } k = 1,2, \ldots N \qquad (3.4)$$

this gives as many equations as there are parameters p_k.

The solution of this set of equations can be represented in a compact form by the use of matrix notation. Let the desired changes in the parameters be given by the vector

$$\Delta p = (\delta p_1, \delta p_2, \delta p_3, \ldots) \qquad (3.5)$$

Let the vector B be defined as:

$$B = \left\{ \sum_i w_i (y_i - \bar{y}_i^0)(\partial \bar{y}_i / \partial p_1), \sum_i w_i (y_i - \bar{y}_i^0)(\partial \bar{y}_i / \partial p_2) \ldots \right\}$$

$$(3.6)$$

The coefficients of the δp_j are represented by a symmetric matrix with the elements

$$C_{lk} = \sum_{i} w(\partial \bar{y}_i / \partial p_l) \cdot (\partial \bar{y}_i / \partial p_k) \qquad (3.7)$$

The set of equations can now be written

$C. \Delta P = B$ where ΔP and B are column vectors.

The solution is $\Delta P = A.B$, where the matrix $A = C^{-1}$.

Since the higher derivatives have been neglected in equation (3.3)this process does not yield the solution of equation(3.2). Therefore, equation(3.4)is solved again with the p_k^0 replaced by $(p_k^0 + \delta p_k)$. This process is repeated in an iterative manner until all the δp_k are less than some convergence criteria. These convergence criteria Δ_k are such that the iterations stop if $p_k \leq \Delta_k$ for all k simultaneously. At this point the fitting process is concluded for this peak.

In order to apply the non linear least squares method to pulse height distributions obtained by means of a Ge(Li) detector system, the following should be considered: A gamma ray spectrum,obtained with a Ge(Li) detector,coupled to a multichannel analyser,consists primarily of a smooth continuous distribution on which is superimposed a series of peaks. Let a portion of this spectrum be represented by the function

$$\bar{y}_i = \sum_j y_{ij} + L_i \qquad (3.8)$$

where

$$y_{ij} = y_{oji} e^{-\mu}\left[1 + \alpha_{1j}(x_i - x_{oj})^{m_1} + \alpha_{2j}(x_i - x_{oj})^{m_2}\right] \qquad (3.9)$$

with

$$\mu = \left[(x_i - x_{oj}) / \left\{\tfrac{1}{2} w_{oj} / (\ln 2)^{1/2}\right\}\right]^2$$

represents the contribution from a single peak and

$$L_i = a + b(x_i - x_1) \qquad (3.11)$$

is a line which represents the continuous distribution.

In a complex peak the subscript j identifies the Gaussian in equation (3.9); the powers m_1 and m_2 are predetermined, even positive integers. The Gaussian parameters x_o (center of photo peak in channels), y_o (count at peak or x_o) w_o (full width of peak at half maximum in channels) can be determined in the fit. The parameters α_1 and α_2, which represent the deviation of the photo peak from a Gaussian, can also be determined. The line parameters a (value at first channel in fit) and b (slope) can be determined in the fit. In some cases the α_1 and α_2 terms have been used in fitting photo peaks from NaI(Tl) data, but a Gaussian function i.e.

$$\alpha_1 = \alpha_2 = 0 \tag{3.12}$$

is usually used for Ge(Li) data.

The correlation technique utilizes the cross correlation function, which is defined as

$$c(\tau)=\lim \frac{1}{2T}\int_{-T}^{+T} f(t)g(t+\tau)dt \quad , \quad \text{when } T \longrightarrow \infty \tag{3.13}$$

where f(t) is the signal to be isolated (e.g. the shape of a peak corresponding to a gamma photon of a certain energy) and g(t) is the signal actually detected (e.g. the pulse height distribution of the complex spectrum). Assuming that

$$g(t) = af(t)+n \tag{3.14}$$

where a is a constant and n represents random noise and approximating f(t) and g(t) with a set of discrete values, the cross correlation function can be approximated as

$$C = \sum_t f_t g_{t+\tau} \tag{3.15}$$

where t and τ are integers (e.g. channel numbers). For the purpose of discussion f(t) is referred to as search spectrum,

$g(t)$ the *data spectrum* and C_τ as the *correlation spectrum*. According to its definition, the cross correlation function is generated by forming the products of $f_{t-\tau}$ and g_t keeping the search function sliding by one unit to the left each time τ is incremented by one. Thus C_τ will have a peak, whenever the peak in the search spectrum overlaps itself in the data spectrum. This fact can be used to localize monoenergetic components in the polyenergetic spectrum. In order to do so successfully, it is necessary to find a method of enhancing the structure in the correlation spectrum; it is also desirable to remove all of the background not associated with the structure. Assuming that the data structure has a form of

$$g_\tau = f_\tau + b \qquad (3.16)$$

with b constant it can be seen that

$$\left.\begin{array}{lll} f_t(g_{t+\tau} -b) \quad 0 & \text{if} & t+\tau < M-1 \\ \text{and} & & \\ f_t(g_{t+\tau} -b) = o & \text{if} & t+\tau > M-1 \end{array}\right\} \qquad (3.17)$$

where M is the number of points in the search spectrum $f(t)$.

This accomplishes both requirements, enhancement of the structure and supression of background. Therefore the expression of C_τ may be modified to read:

$$C_\tau = \sum_{t=0}^{M-1} f_t \left[g_{t+\tau} - A_\tau \right]$$

where (3.18)

$$A_\tau = \frac{1}{M} \sum_{n=\tau}^{\tau+M-1} g_n$$

When applying this method to actual data, certain modifications have to be introduced in order to take care on random fluctuations. It is clear that the term

$$g_{t+\tau} - A_\tau$$

will no longer vanish beyond the structure for data where A is fluctuating in a statistical manner. However, remembering that the computation of A_τ , as given by equation (3.18), it can certainly be expected that fluctuations of one standard deviation or more would be smoothed out. Therefore the square root of A should be sufficient to suppress statistical fluctuations. With this in mind, the modified equation for C now reads:

$$C_\tau = \sum_{t=0}^{M-1} f_t \left[g_{t+\tau} - A + \sqrt{A} \right] \qquad (3.19)$$

When applying the cross correlation method to decomposition of pulse height spectra, the search spectrum is a Gaussian, therefore

$$C_\tau = N_0 \sum_{t=0}^{M-1} \exp \left[-\left(\frac{t-t_0}{b}\right)^2 \right] \left[g_{t+\tau} - A_\tau + \sqrt{A_\tau} \right] \qquad (3.20)$$

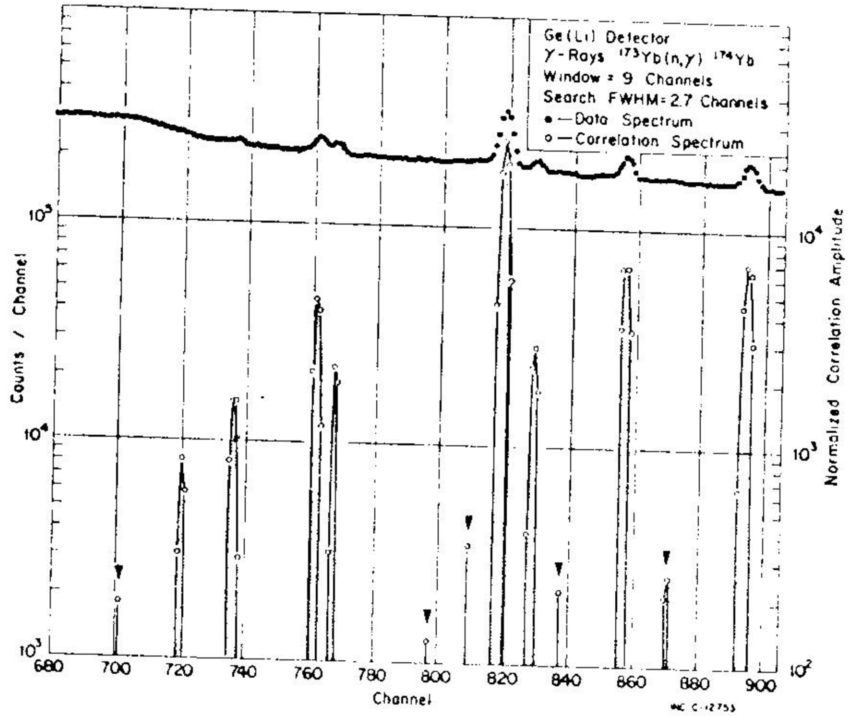
where

$$\mathrm{FWHM} = 2b\sqrt{\ln 2} \qquad (3.21)$$

The only input parameters are the window width M and the FWHM. N_o is a constant.

Figures 3.1 and 3.2 contain the pulse height distributions (data spectrum) and the pertinent cross correlation functions obtained using appropriate search spectra, plotted on the same diagram. Figure (3.1) refers to a spectrum obtained by a Ge(Li) detector. Figure (3.2) to one obtained by a NaI(Tl) crystal.

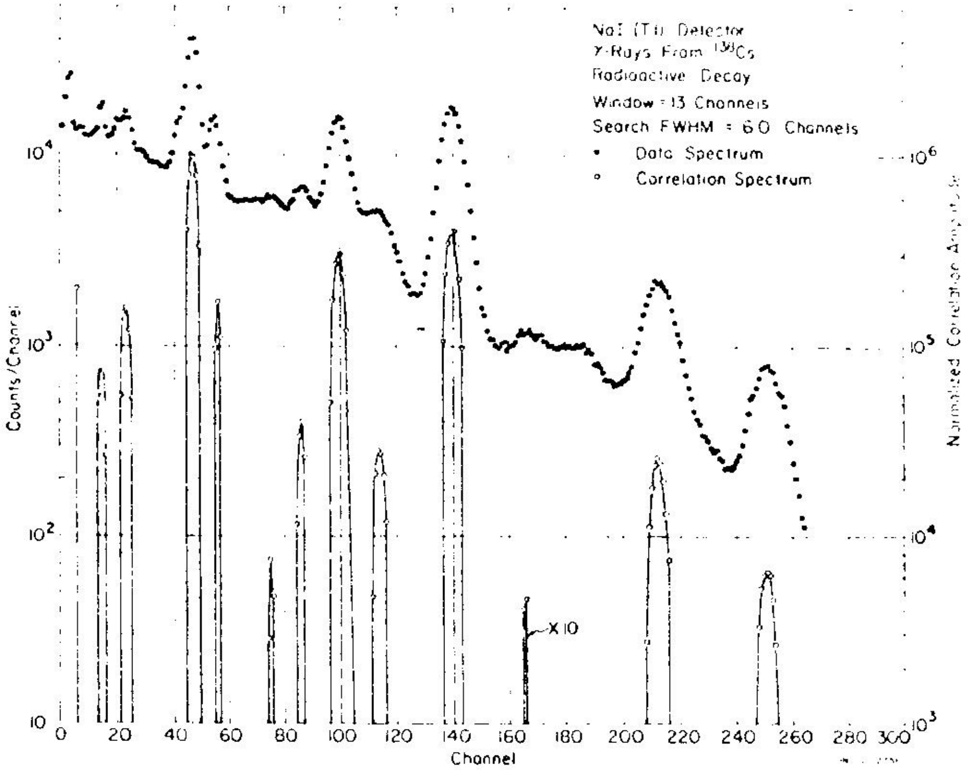
Both figures (quoted from reference 11) illustrate the effectiveness of the cross correlation method in locating peaks.



Data spectrum obtained with a lithium-drifted germanium detector. These data are exemplary of neutron-capture gamma-ray experiments.

FIGURE 3.1

(from reference 11)



Results of applying the correlation method to data obtained with a sodium-iodide detector. This example illustrates the insensitivity of the method to the peak-width parameter, and also how Compton edges and backscatter peaks can result in structure appearing in the correlation spectrum.

FIGURE 3.2

(from reference 11)

CHAPTER IV

MATHEMATICAL BASIS OF THE PRGA-CODE

The PRGA code is based on a mathematical model using linear least squares techniques to calculate the mixing factors of a multicomponent spectrum.

In its simplest form, the problem may be formulated in the following manner:

Given the pulse height distribution spectrum of a source mixture, obtained by means of a certain gamma ray detector system, as well as normalized pulse height distribution spectra of the individual components obtained by means of the same gamma ray detector system, the mixing factors are to be determined.

In order to obtain a numerical solution of this problem, the following mathematical model is used:

Let S(I,K) denote the count rate in the K th channel due to the I th isotope

C(I) the I th mixing factor to be determined

T(K) the count rate in the K th channel due to mixture (given)

II the total number of isotopes used in the mixture (given)

KK the total number of channel used (given)

Under the assumption that T(K) the count rate in the

K th channel due to the source mixture is produced by a linear combination of count rates corresponding to each component, T(K) may be expressed as:

$$T(K) = \sum_{I=1}^{II} C(I) * S(I,K) \qquad \text{for } K=1,2, \ldots KK \qquad (4.1)$$

In order to determine the optimal values of the C(I) mixing factors a linear least squares fit is used, searching for a minimum of the function:

$$U(C_1, C_2, \ldots C_{II}) = \sum_{K=1}^{KK} \left[T(K) - \left\{ \sum_{I=1}^{II} C(I) * S(I,K) \right\} \right]^2 \qquad (4.2)$$

with respect of the choice of the C(I) mixing factors. The minimizing C(I) values are determined by differentiating U with respect to each C(I) and setting the derivatives to zero. This leads to the system of linear equations of the following form:

$$\frac{\partial U}{\partial [C(J)]} = - \sum_{K=1}^{KK} \left\{ \left[T(K) - \sum_{I=1}^{II} C(I) * S(I,K) \right] S(J,K) \right\} = 0 \qquad (4.3)$$

leading to :

$$\sum_{K=1}^{KK} T(K) * S(J,K) = \sum_{I=1}^{II} C(J) \left[\sum_{K=1}^{KK} S(I,K) * S(J,K) \right]$$

$$\text{for } J = 1, 2, \ldots II \qquad (4.5)$$

denoting

$$\sum_{K=1}^{KK} T(K)*S(J,K) = B(J) \qquad (4.6)$$

and

$$\sum_{K=1}^{KK} S(I,K)*S(J,K) = A(I,J) \qquad (4.7)$$

one obtains

$$\sum_{I=1}^{II} A(I,J)*C(I) = B(J) \qquad (4.8)$$

for J=1,2, ...II

The solution of the set of this system of equations yields the desired C(I) values.

To execute the above computation of the C(I) values, a computer program PRGA was developed. The arrangement of the card deck and the listing of the program are presented in APPENDIX I.

PRGA is written in FORTRAN IV, level 19, and is operative on the IBM 360/40 and on the DEC-PDP-10, both at the University of Puerto Rico, Mayaguez Campus, Computer Center.

In order to facilitate the debugging procedure, the PRGA was first tested on a simulated problem, which assumed a source mixture consisting of ten components and data from forty one channels.

After its performance on the simulated problem has been found excellent, PRGA was applied to actual experimental data, and the computed results found to be in satisfactory agreement with premeasured control data.

A brief description of the experimental equipment and procedures as well as a detailed comparison of the experimental mixing factors with those computed by the PRGA program are given in Chapters V and VI.

The performance of the PRGA has been improved by the inclusion of several routines for various corrections. The mathematical basis of these routines is briefly presented below:

a. Dead time correction was taken into account by first including the system dead time as an input parameter then correcting each raw count rate reading S in the following way:

$$S = \frac{S}{1 - \tau S} \qquad (4.9)$$

where S is the raw count rate per second and is the system dead time in seconds. Dead time values used in this paper were computed from the percent values automatically recorded by the counting system. No attempt was made to include channel and half life dependent dead time correction such as advocated by W. FILIPPONE and F. J. MUNNO (see ref. 22) in the code, at this stage of its development, primarily because all control data referred to sources with half lives of several tens of days, thus the advocated correction would be negligible.

b. Correction for natural background ia made simply by including the appropriate background count rates BB(K) as inputs in the program and by replacing

$$S(I,K) \longleftarrow S(I,K)-BB(K) \qquad (4.10)$$

$$\text{for } K = 1,2, \ldots KK$$

c. Corrections related to counting statistics. Due the inherently random nature of radio active disintegration, allowance had to be made for the statistical fluctuations in the count rates. This was accomplished by assigning a weight factor w_k to each value of

$$\left[T(K) - \sum_{I=1}^{II} S(I,K)*C(I)\right]$$

modifying the function entering the least squares fit in the following manner:

$$U = \sum_{K=1}^{KK} w_k \left[T(K) - \sum_{I=1}^{II} S(I,K)*C(I)\right]^2 \qquad (4.11)$$

d. Correction for Compton scattering was made in a purely heuristic fashion, by deducting the pertinent ordinates of an appropriately chosen linear function from the measured values. Assuming that the peak occurs at $K=K_o$ and the tangential points S at K_o-M

resp. K_o+M the linear function to be deducted is given by

$$Y(K) = \frac{Y(K_o+M)-Y(K_o-M)}{2M}(K-K_o+M)+Y(K_o-M) \qquad (4.12)$$

and the proper correction consists in replacing

$S(I,K) \longleftarrow S(I,K)-Y(K)$ for

$$K = K_o-M,\ K_o-M+1,\ \ldots,\ K_o+M \qquad (4.13)$$

Admittedly, the nonlinear method of fitting the individual peaks to the sum of a Gaussian and a linear function, as described in Chapter III, is better justified from the theoretical point of view; it should be observed, however, that even the above described simple linear routine is used by several authors (refs. 5, 9) and that its use in PRGA yielded satisfactory results.

e. Decay corrections are necessary when the code is applied in neutron activation analysis involving short lived isotopes.

In order to understand how this correction has been applied let us consider the time behaviour of a single isotope's activity assuming that

0 to t_1 - is the irradiation time
t_1 to t_2 - is the cooling time
t_2 to t_3 - is the counting interval

Φ - the activating flux

λ_i - is the decay constant of the i-th isotope

σ_i - is the activation (microscopic) cross section of the nuclide producing the i-th isotope

$A_i(t)$ - the time dependent activity of the i-th isotope

$N_i(t)$ - the time dependent number density of the i-th isotope and

N_o^i - the number density of the nuclide, the activation of which yields the i-th isotope.

Under these assumptions the following holds:

$$\frac{dN_i(t)}{dt} = N_o^i\Phi\sigma_i - \lambda_i N_i(t) \qquad 0 < t < t_1 \qquad (4.14)$$

leading to

$$N_i(t) = \frac{N_o^i\Phi\sigma_i}{\lambda_i}\left[1-e^{-\lambda_i t}\right] \qquad 0 < t < t_1 \qquad (4.15)$$

during cooling, i.e. when $t_1 < t < t_2$

$$N_i(t) = \frac{N_o^i\Phi\sigma_i}{\lambda_i}\left[1-e^{-\lambda_i t_1}\right]\left[e^{-\lambda_i(t-t_1)}\right] \qquad (4.16)$$

and during counting, i.e. when $t_2 < t < t_3$

$$N_i(t) = \frac{N_o^i\Phi\sigma_i}{\lambda_i}\left[1-e^{-\lambda_i t_1}\right]\left[e^{-\lambda_i(t-t_1)}\right] \qquad (4.17)$$

The activity of the sample at any instant of the counting interval is given by: $t_2\ t\ t_3$

$$A_i(t)=\lambda_i N_i(t)=N_o^i\sigma_i\Phi\,(1-e^{-\lambda_i t_1})e^{-\lambda_i(t-t_1)} \tag{4.18}$$

Therefore the count rate due to the i-th isotope will be proportional to:

$$\int_{t_2}^{t_3} A_i(t)dt=N_o^i\sigma_i\left[1-e^{-\lambda_i t_1}\right].\Phi.e^{-\lambda_i t_1}\int_{t_2}^{t_3} e^{-\lambda_i t}dt = \frac{N_i^o\,\sigma_i\Phi}{\lambda_i}\left[1-e^{-\lambda_i t_1}\right]\left[e^{-\lambda_i(t_2-t_1)}-e^{-\lambda_i(t_3-t_1)}\right] \tag{4.19}$$

Hence in the case of a mixture consisting of short lived isotopes the mixing factors that will result from the least squares fit - described above - will depend upon t_1, t_2, t_3, σ_i, and Φ and in order to get mixing factors characterizing the composition before irradiation, the following correction has to be applied:

$$CC(I)=\frac{C(I)\,\lambda_i}{\sigma_i\Phi\left[1-e^{\lambda_i t_1}\right]\left[e^{-\lambda_i(t_2-t_1)}-e^{-\lambda_i(t_3-t_1)}\right]} \tag{4.20}$$

From the mixing factors, CC(I), of the unirradiated sample, the actual weights are computed by using the normalized activities and weights of the corresponding standard samples.

In order to compute the weights of the individual components in the unirradiated mixture, the following basic equation has to be used:

$$T(K) = \Phi \sum_{J=1}^{II} W(J) * A(K,J) * \left[\frac{e^{-\lambda_j (t_2 - t_1)} - e^{-\lambda_j (t_2 - t_4)}}{\lambda_j} \right] \left[1 - e^{-\lambda_j t_1} \right] \qquad (4.21)$$

where:

T(K) = observed counts in channel K

Φ = neutronflux

W(J) = weight of the J-th isotope in the sample

A(K,J) = specific activity for radioisotope J at channel K

λ_j = decay factor for radio isotope J.

The validity of the above equation is subject to the following restrictions:

(a) The number of counts observed for any component is directly proportional to the amount of the component present.

(b) The spectra is additive.

(c) The flux remains constant during irradiation.

(d) The "deadtime" of the data acquisition system does not change during any counting period.

This results in a set of equations equal in number to the number of channels. If the specific activities A(K,J) have been determined by prior calibration, the this set of equations can be solved for the unknown weights W(J) using least square techniques.

CHAPTER V

EXPERIMENTAL PROCEDURES AND EQUIPMENT

The performance of the PRGA code was evaluated by comparing premeasured mixing factors with those computed by the code.

Standard sources and source mixtures were prepared from a set of liquid reference sources manufactured by Atomic Development Corp.. The set included the following nuclides:

NUCLIDE	MOST PROBABLE ENERGIES MeV	HALF LIFE
Na^{22}	0.511, 1.275	2.62 yr.
Mn^{54}	0.829	300 days
Co^{57}	0.122, 0.136	270 days
Co^{60}	1.173, 1.332	5.2 yr.
Cs^{137}	0.662	30 yr.

In order to ensure adequate accuracy self filling lambda pipettes were used for measuring radioactive liquids.

Health Physics regulations and instructions referring to handling open sources were strictly observed. After receiving the reference sources, they were tested for leakage by the

PRNC - Health Physics Division (see copy of leak test report). All operations involving liquid sources were performed under a hood, with the experimenter wearing protective gloves.

Standards and mixtures were prepared at the same time in order to avoid errors due to decay. An aliquot of the reference liquid (e.g. 500 λ) was pipetted on a planchet. A multiple of that aliquot was entered into the planchet that was designated to hold the mixture. Care was taken to deposit the liquid at the center of the planchet in order to preserve identical geometry as much as possible. All sources were dried under a lamp and after drying all were covered with thin plastic covers.

All sources were counted on the Ge(Li) spectrometer of the PRNC Radioecology Division.

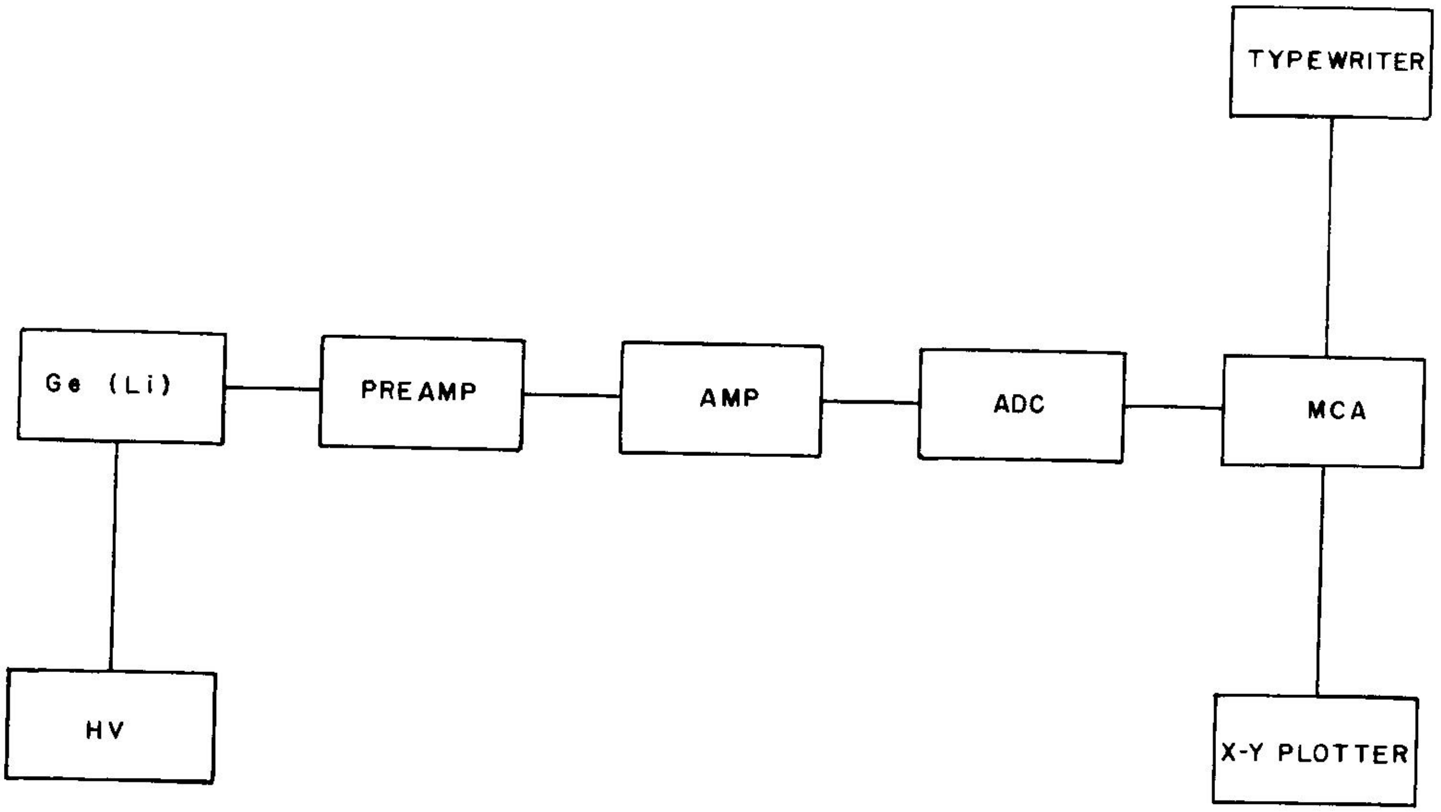
A block diagram of the counting equipment is shown in Figure 5.1 and a list of the components is given below.

Ge(Li) detector and associated data acquisition system components:

HV - High voltage power supply, manufactured by Hewlett-Packard; model No. 3015.

Ge(Li) - Lithium drifted germanium detector, manufactured by Canberra Industries Co.; model No. 7229-340; with cryostat model No. 7500.

PRE AMP - Preamplifier, manufactured by Canberra



BLOCK DIAGRAM OF Ge (Li) DETECTOR AND ASSOCIATED DATA ACQUISITION SYSTEM

FIGURE 5.1

Industries Co.; model No. 970.

AMP - Main Amplifier, manufactured by Tennelec; model No. TC200.

ADC - Analog to digital converter manufactured by Hewlett-Packard, model No. 160.

MCA - Multichannel Analyzer, manufactured by Hewlett-Packard; model No. 45.

TYPEWRITER Manufactured by IBM; model No. 11-C.

X-Y PLOTTER Manufactured by Hewlett-Packard; model No. 7590C.

The Ge(Li) detector used in this system is of open ended coaxial pin type. The starting material for the detector is p type germanium (doped with gallium). Lithium, acting as a donor is diffused, at high temperature (400 C°) into the cylindrical ingot creating a thin n-type layer on the surface. A bias voltage is used to cause the lithium ions to drift through the p type material. During this process an equilibrium condition is established where lithium ions pair with atoms of the doping material (gallium) creating a region with intrinsic properties. (See Figure 5.2).

The term intrinsic implies that charge carriers will have a very long lifetime with all recombination processes reduced to a negligible level. Under reverse bias the charge carriers can then be collected from this region resulting in the production of a solid state ionization cham-

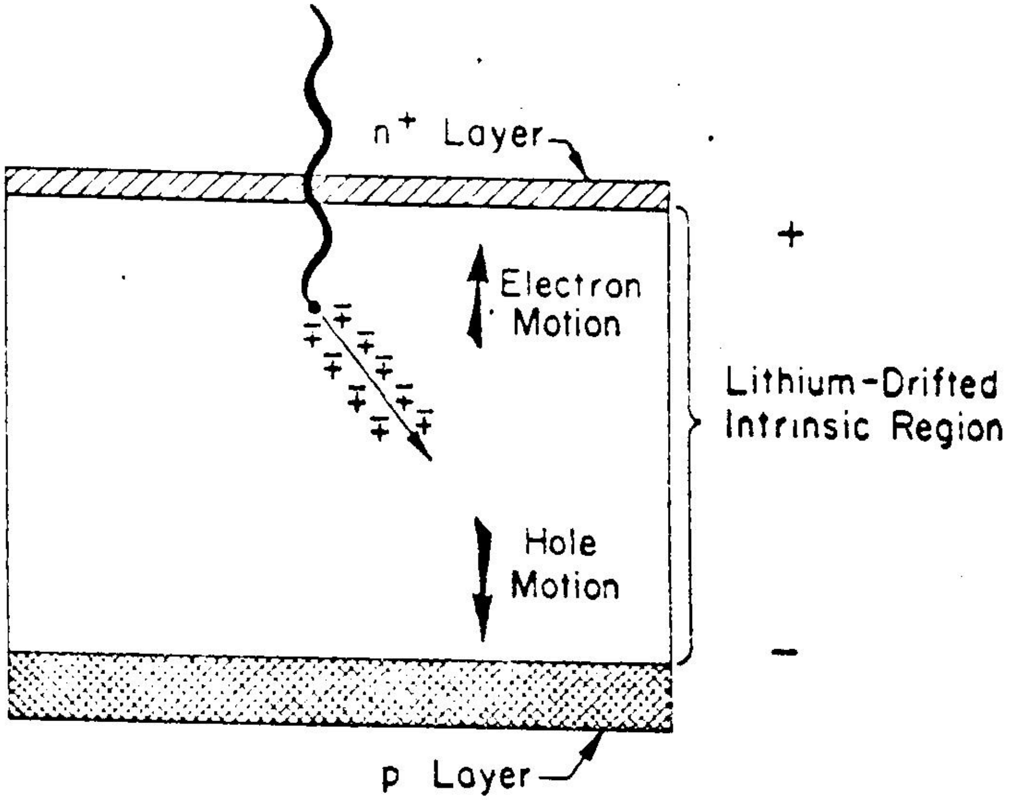


FIGURE 5.2

(from reference 28)

ber, schematically represented in Figure 5.2.

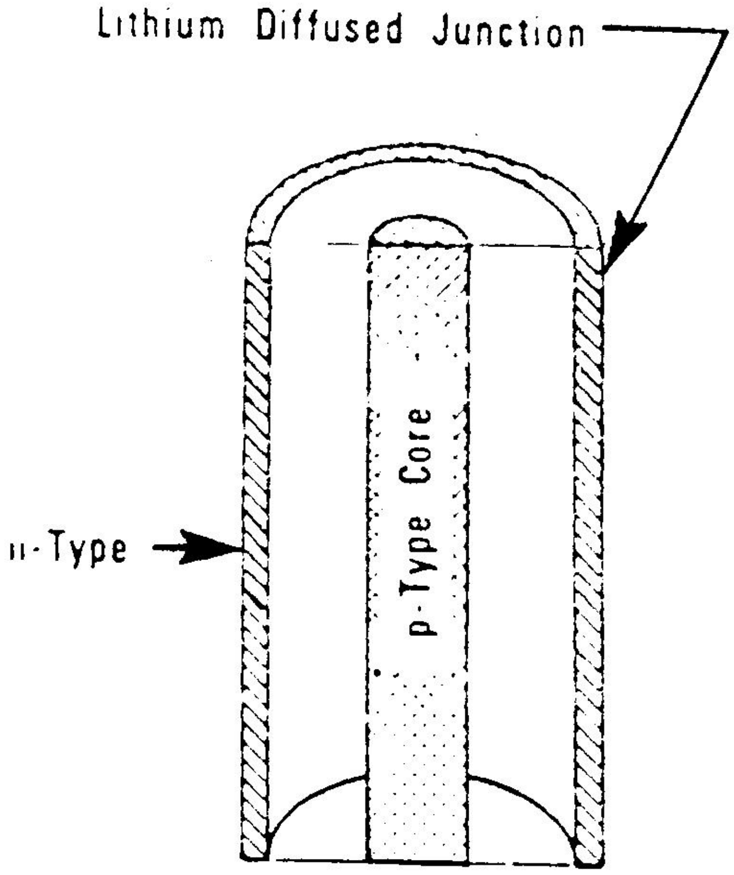
Since the mobility of lithium ions in germanium is not negligible at room temperature, such devices once fabricated, must be maintained at low temperatures to preserve the structure. A liquid nitrogen filled cryostate was used to maintain the required temperatures.

Figure 5.3 indicates the configuration of the Ge(Li) detector actually used in this project.

On the average only 2.84 eV is required to create an electron-hole pair in a Ge(Li) detector as compared to 30 eV in a gas filled ionization chamber or to 500 eV required to produce a photoelectron at the photo cathode of a scintillation detector system. It is this reduced energy requirement that improves the statistics and thereby the resolution of Ge(Li) detectors in comparison to gas-ionization chambers or scintillation detectors.

In Figure 5.4 energy vs. channel number is plotted, indicating excellent linearity of the counting system used for data acquisition. Source mixtures and pertinent standards were counted within a few hours - thus reducing errors due to decay. Maximum counting registered by the inbuilt deadtime counter were below 2%. Within each counting session the gainshift remained below observable.

In order to ensure acceptable counting statistics one hour counts were taken. Countrate-outputs were obtained both on an IBM typewriter and on an X-Y plotter.



Open-ended Coaxial Ge(Li) Detector

FIGURE 5.3

(from reference 28)

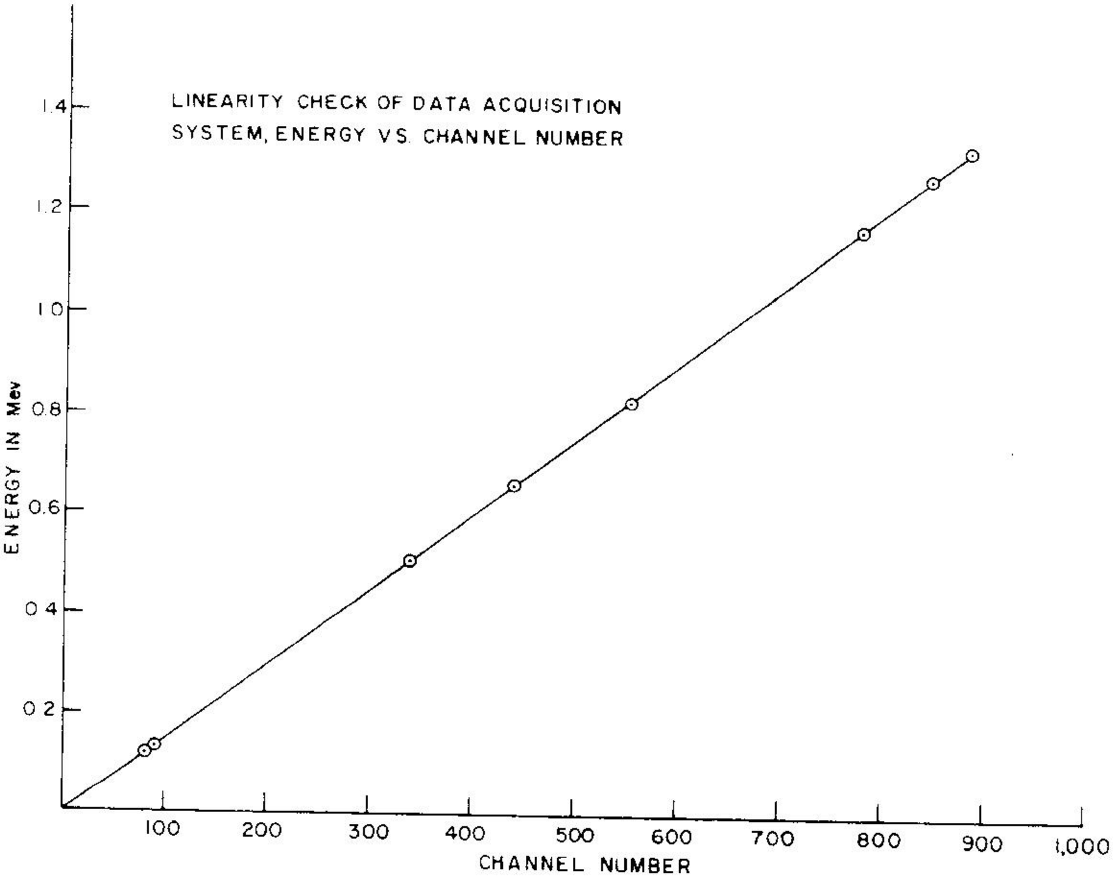

LINEARITY CHECK OF DATA ACQUISITION
SYSTEM, ENERGY VS. CHANNEL NUMBER
ENERGY IN Mev
1.4
1.2
1.0
0.8
0.6
0.4
0.2
100
200
300
400
500
600
700
800
900
1,000
CHANNEL NUMBER

FIGURE 5.4

CHAPTER VI

PRESENTATION AND DISCUSSION OF RESULTS

PRGA computed results are presented in tabulated form side by side with premeasured mixing factors in order to facilitate comparison of the two. Entries in Table 6.1 refer to the ten-component simulated problem. Tables 6.2 through 6.5 contain results referring to two, three, four and five component source mixtures respectively. Discrepancies of premeasured and PRGA computed mixing factors are indicated in percents. Figures 6.1 through 6.8 are samples of pulse height distribution spectra of standard sources and of source mixtures plotted from data obtained by the Ge(Li) detector equipped data acquisition system described in Chapter V.

The excellent agreement between assigned and PRGA computed mixing factors in Table 6.1 indicate that the performance of the PRGA-code is essentially right. When applied to experimentally acquired data, which by the nature of the acquisition are affected by such factors as gainshift, counting statistics, spectral background, etc., the discrepancy of premeasured and PRGA computed results - understandably enough - increase substantially.

The PRGA computed results presented in Tables 6.2 through 6.5 are including corrections intended to compensate for errors related to background, deadtime, Compton scattering and counting statistics.

Comparable published results quoted from three references are presented in Tables 6.6 through 6.8.

Based upon the results presented above, the following can be concluded:

(a) The code developed can determine mixing factors of source mixtures, using gamma spectrum of the composite source as well as those of the standard sources as input.

(b) The PRGA-code is operative on the IBM/360-40 and the DEC-10 computer both at the Computer Center of the University of Puerto Rico, Mayaguez Campus.

(c) PRGA's performance on a ten component simulated problem was found excellent.

(d) PRGA computed mixing factors have been found in satisfactory agreement with premeasured values in a set of measurements performed on two, three, four and five component mixtures - especially prepared from reference source liquids for the purpose of this test.

(e) As indicated by entry 3 in Table 6.3,PRGA gives a near-zero mixing factor for a standard absent from the composite spectrum.

TABLE 6.1

COMPARISON OF ASSIGNED AND PRGA-COMPUTED MIXING FACTORS FOR TEN COMPONENT SIMULATED PROBLEM

Component	MIXING FACTORS		Discrepancy (%)
	Assigned	PRGA Computed	
A	5.20	5.20	0.00
B	7.30	7.19	1.51
C	4.50	4.51	0.22
D	6.80	6.81	0.15
E	10.60	10.61	0.09
F	13.80	13.81	0.07
G	11.40	11.40	0.00
H	0.86	0.85	1.16
I	3.80	3.80	0.00
K	0.45	0.45	0.00

TABLE 6.2

COMPARISON OF PREMEASURED AND PRGA-COMPUTED MIXING FACTORS FOR TWO COMPONENT SOURCE MIXTURES

SAMPLE NUMBER	COMPONENT	MIXING FACTORS PRGA-COMP.	PREMEAS.	DISCREPANCY (%)
1	$11\mathrm{Na}^{22}$	1.10	1.00	10.0
	$55\mathrm{Cs}^{137}$	1.08	1.00	8.0
2	$27\mathrm{Co}^{57}$	2.35	2.5	6.0
	$11\mathrm{Na}^{22}$	3.27	3.33	1.8
3	$27\mathrm{Co}^{57}$	10.0	8.47	15.3
	$11\mathrm{Na}^{22}$	7.0	5.90	15.7

TABLE 6.3

COMPARISON OF PREMEASURED AND PRGA-COMPUTED MIXING FACTORS FOR THREE COMPONENT SOURCE MIXTURES

SAMPLE NUMBER	COMPONENT	MIXING FACTORS		DISCREPANCY %
		PREMEAS.	PRGA-COMP.	
1	${}_{11}\mathrm{Na}^{22}$	2	1.86	7.0
	${}_{27}\mathrm{Co}^{57}$	1	0.90	10.0
	${}_{25}\mathrm{Mn}^{54}$	2	1.99	0.5
2	${}_{11}\mathrm{Na}^{22}$	0.33	0.32	3.0
	${}_{27}\mathrm{Co}^{57}$	5.00	5.12	2.4
	${}_{27}\mathrm{Co}^{60}$	0.15	0.17	13.3
3	${}_{11}\mathrm{Na}^{22}$	3.33	3.09	7.2
	${}_{27}\mathrm{Co}^{57}$	2.5	2.37	5.2
	${}_{27}\mathrm{Co}^{60}$	0	0.07	-

TABLE 6.4

COMPARISON OF PREMEASURED AND PRGA-COMPUTED MIXING FACTORS FOR FOUR COMPONENT SOURCE MIXTURES

SAMPLE NUMBER	COMPONENT	MIXING FACTORS		DISCREPANCY %
		PREMEAS.	PRGA-COMP.	
1*	$_{11}Na^{22}$	2	1.62	19.0
	$_{25}Mn^{54}$	1	0.88	12.0
	$_{27}Co^{60}$	2	1.95	2.5
	$_{55}Cs^{137}$	2	1.64	18.0
2**	$_{11}Na^{22}$	2	1.64	18.0
	$_{25}Mn^{54}$	1	0.92	8.0
	$_{27}Co^{60}$	2	1.95	2.5
	$_{55}Cs^{137}$	2	1.61	19.5

* Computed with w_k = 1/T(K)

** Computed with w_k = 1/SQRT(T(K))

TABLE 6.5

COMPARISON OF PREMEASURED AND PRGA-COMPUTED MIXING FACTORS FOR FIVE COMPONENT SOURCE MIXTURES

SAMPLE NUMBER	COMPONENT	MIXING FACTORS		DISCREPANCY %
		PREMEAS.	PRGA-COMP.	
1*	$_{11}Na^{22}$	1	0.92	8.00
	$_{27}Co^{57}$	1	0.76	24.00
	$_{25}Mn^{54}$	1.6	1.45	9.37
	$_{27}Co^{60}$	1	1.03	3.00
	$_{55}Cs^{137}$	2	1.81	9.50
2**	$_{11}Na^{22}$	1	0.93	7.00
	$_{27}Co^{57}$	1	0.79	21.00
	$_{25}Mn^{54}$	1.6	1.47	8.10
	$_{27}Co^{60}$	1	1.03	3.00
	$_{55}Cs^{137}$	2	1.77	11.50

* Computed with w_k = 1/T(K)

** Computed with w_k = 1/SQRT(T(K))

TABLE 6.6

RESULTS QUOTED FROM REFERENCE (25)
FOR TEN COMPONENT MIXTURE

COMPONENT	QUANTITY		DISCREPANCY %
	PREMEASURED	COMPUTED	
11Na^{22}	5.00 μgm	7.7 μgm	54.0
17Cl^{38}	36.8 μgm	38.0 μgm	3.3
19K^{42}	0.404 mgm	0.301 mgm	25.4
25Mn^{56}	0.101 μgm	0.0904 μgm	10.4
21Sc^{46}	14.2 μgm	12.2 μgm	14.1
33As^{76}	5.00 μgm	4.98 μgm	0.4
29Cu^{64}	2.23 μgm	2.11 μgm	0.9
24Cr^{51}	1.82 gm	1.56 μgm	14.0
53I^{128}	2.49 μgm	2.68 μgm	7.6
57La^{140}	2.19 μgm	3.02 μgm	37.9

TABLE 6.7

RESULTS QUOTED FROM REFERENCE (8) FOR THREE COMPONENT SOURCE MIXTURES

COMPONENT	MIXING FACTORS		DISCREPANCY %
	PREMEASURED	COMPUTED	
$_{58}Ce^{144}$	1	1.03	2.91
$_{55}Cs^{137}$	5.2	5.31	2.07
$_{27}Co^{60}$	5	5.11	2.15

TABLE 6.8

RESULTS QUOTED FROM REFERENCE (6) FOR FIVE COMPONENT MIXTURE

COMPONENT	MIXING FACTORS		DISCREPANCY %
	PREMEASURED	COMPUTED	
1	1	0.990	1.00
2	1	1.009	0.89
3	1	1.008	0.79
4	1	1.015	1.50
5	1	1.010	0.99

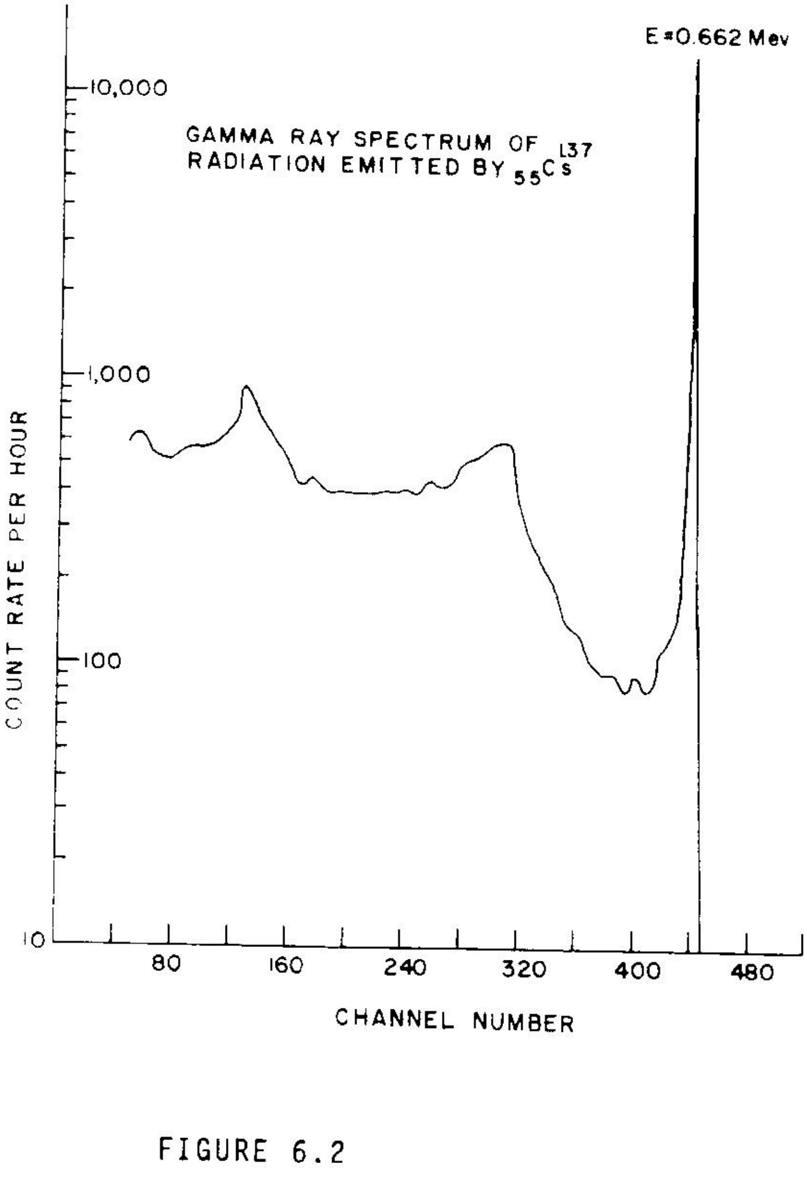

E=0.662 Mev
10,000
GAMMA RAY SPECTRUM OF RADIATION EMITTED BY $_{55}Cs^{137}$
1,000
100
10
COUNT RATE PER HOUR
80
160
240
320
400
480
CHANNEL NUMBER

FIGURE 6.2

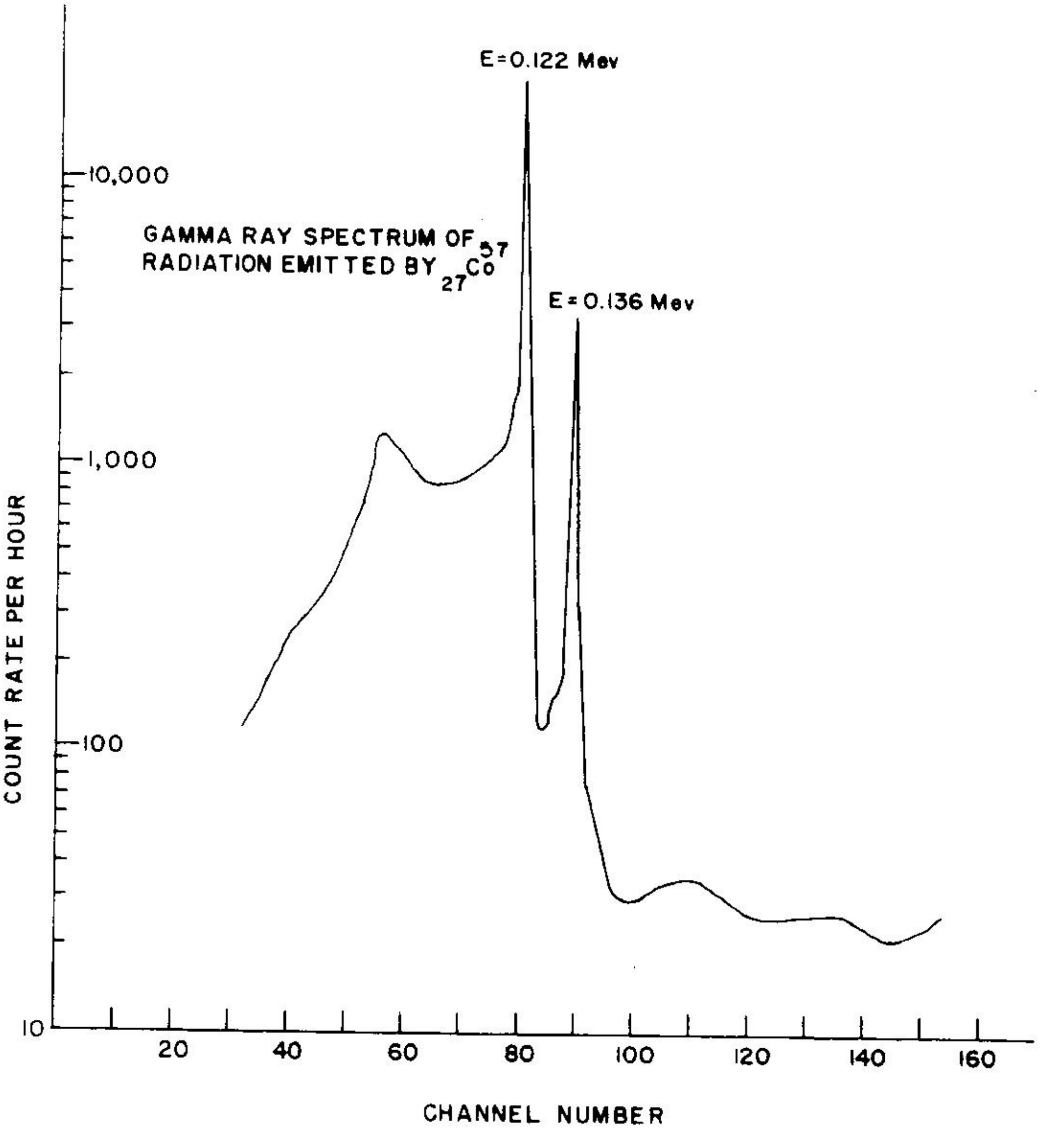

E=0.122 Mev
E=0.136 Mev
GAMMA RAY SPECTRUM OF $_{27}Co^{57}$
RADIATION EMITTED BY
10,000
1,000
100
10
COUNT RATE PER HOUR
20
40
60
80
100
120
140
160
CHANNEL NUMBER

FIGURE 6.3

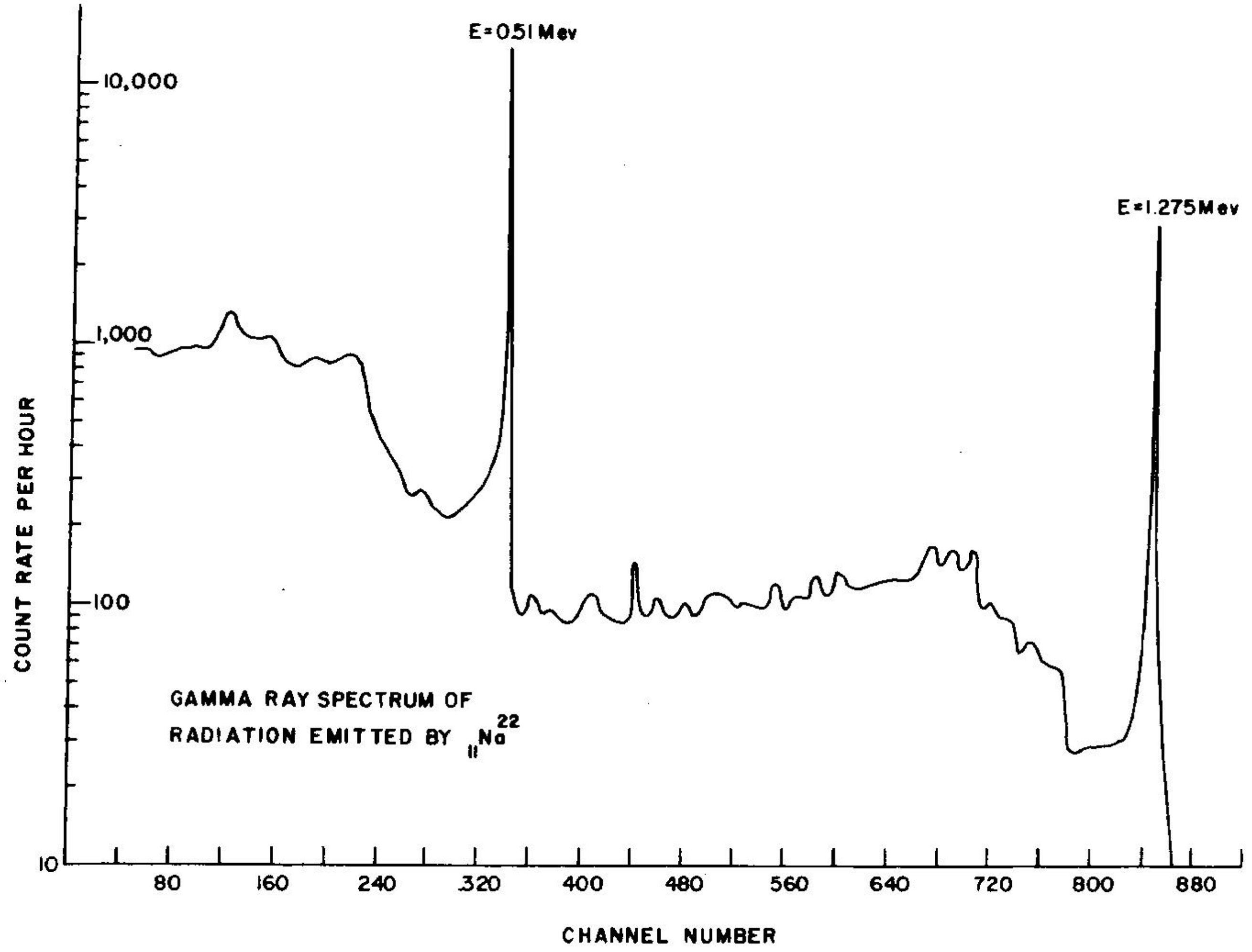

E=0.51Mev
E=1.275Mev
10,000
1,000
100
10
COUNT RATE PER HOUR
GAMMA RAY SPECTRUM OF
RADIATION EMITTED BY $_{11}Na^{22}$
80
160
240
320
400
480
560
640
720
800
880
CHANNEL NUMBER

FIGURE 6.4

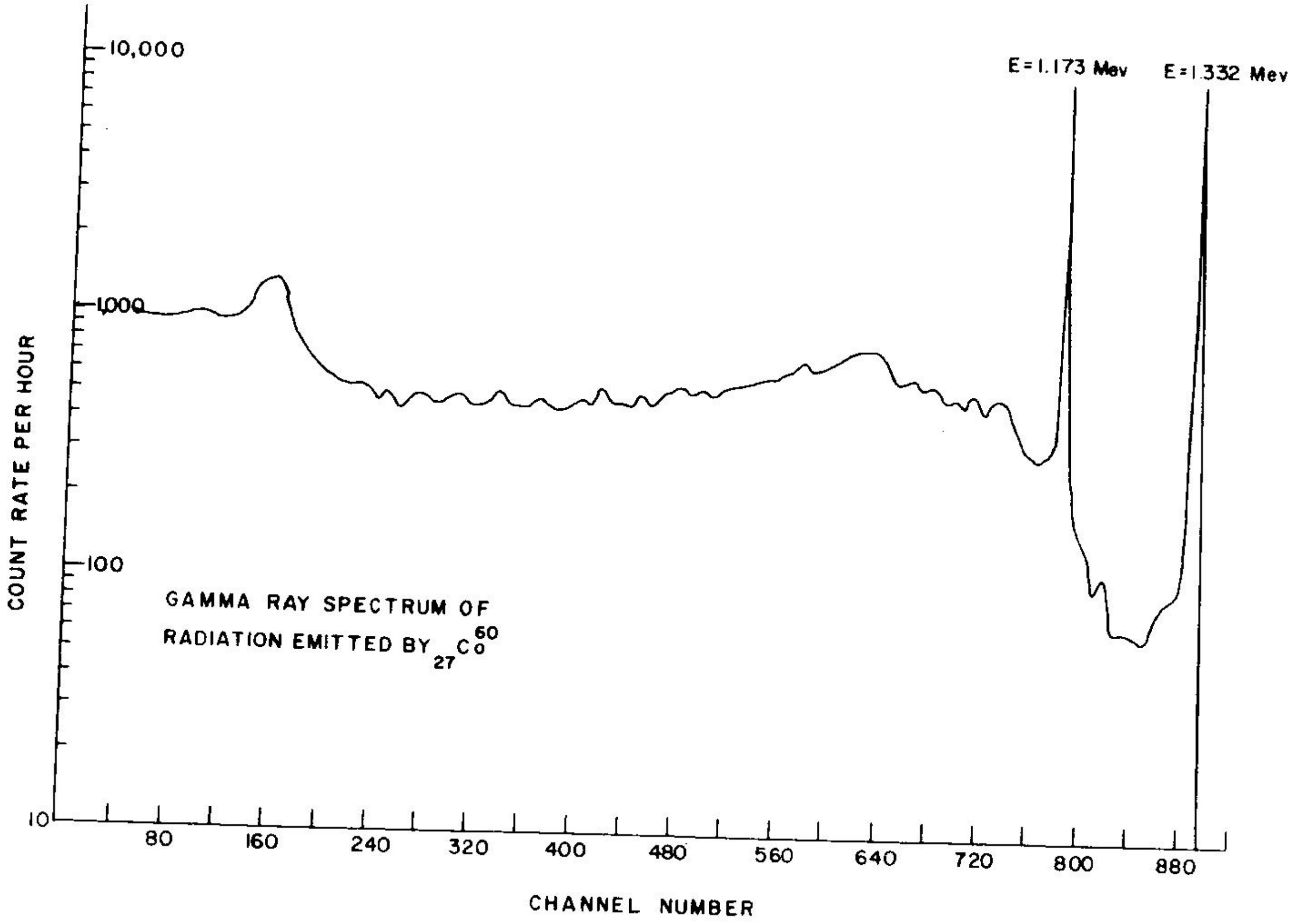

E=1.173 Mev
E=1.332 Mev
10,000
1,000
100
10
COUNT RATE PER HOUR
GAMMA RAY SPECTRUM OF
RADIATION EMITTED BY $_{27}Co^{60}$
80
160
240
320
400
480
560
640
720
800
880
CHANNEL NUMBER

FIGURE 6.5

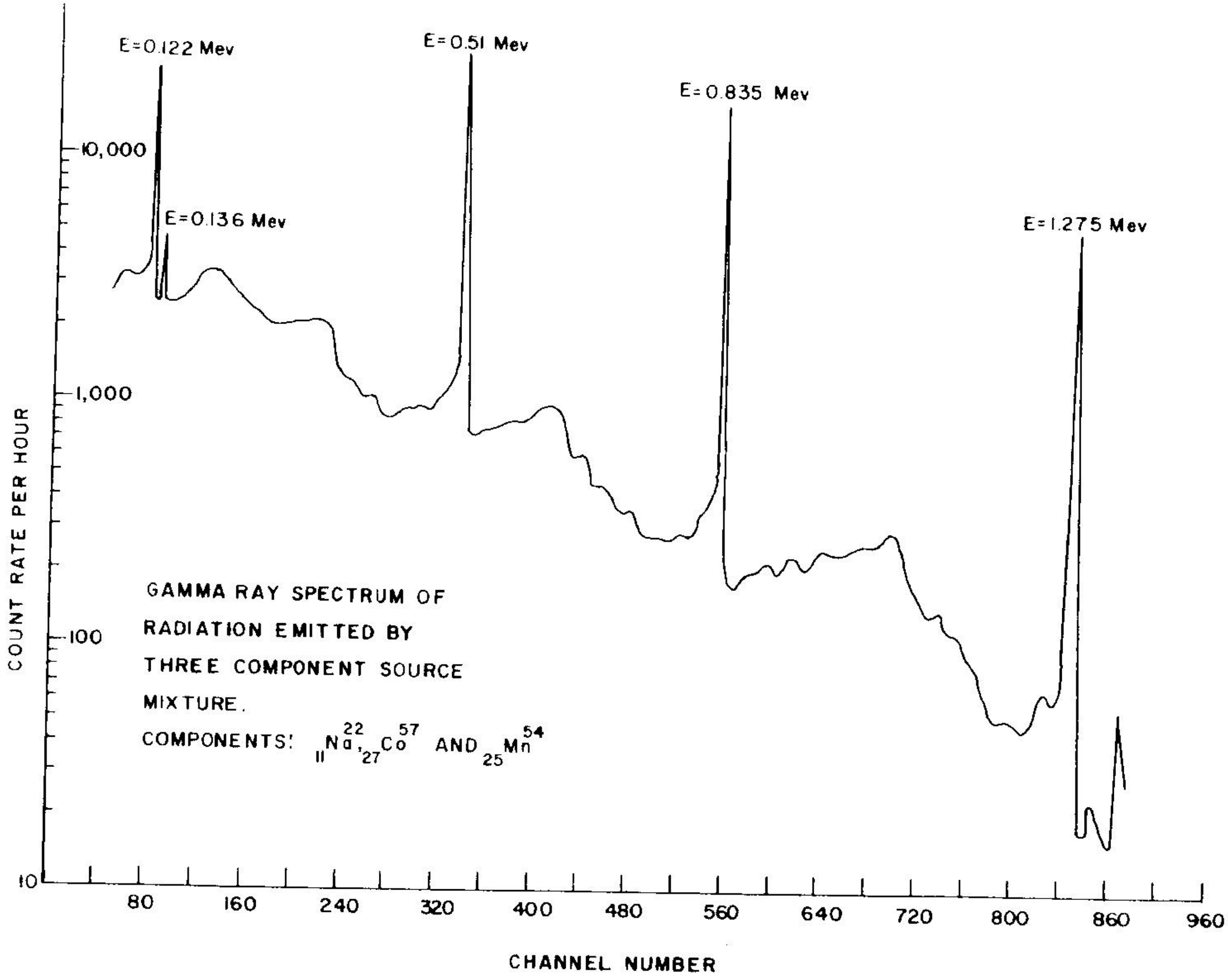

E=0.122 Mev
E=0.136 Mev
E=0.51 Mev
E=0.835 Mev
E=1.275 Mev
10,000
1,000
100
10
COUNT RATE PER HOUR
80
160
240
320
400
480
560
640
720
800
860
960
CHANNEL NUMBER
GAMMA RAY SPECTRUM OF
RADIATION EMITTED BY
THREE COMPONENT SOURCE
MIXTURE.
COMPONENTS: ${}_{11}Na^{22}$, ${}_{27}Co^{57}$ AND ${}_{25}Mn^{54}$

FIGURE 6.6

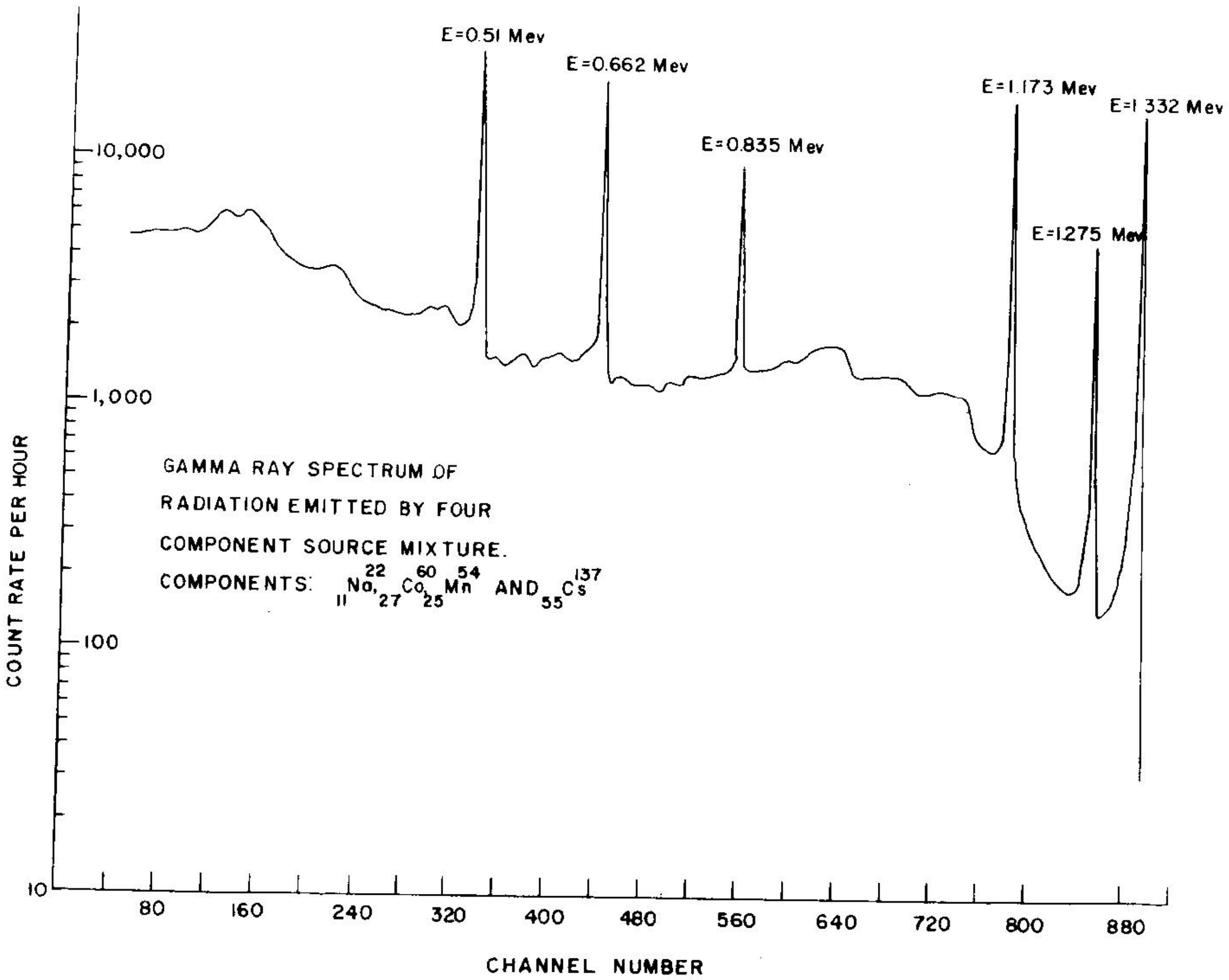

E=0.51 Mev
E=0.662 Mev
E=0.835 Mev
E=1.173 Mev
E=1.275 Mev
E=1.332 Mev
10,000
1,000
100
10
COUNT RATE PER HOUR
GAMMA RAY SPECTRUM OF
RADIATION EMITTED BY FOUR
COMPONENT SOURCE MIXTURE.
COMPONENTS: $_{11}Na^{22}$, $_{27}Co^{60}$, $_{25}Mn^{54}$ AND $_{55}Cs^{137}$
80
160
240
320
400
480
560
640
720
800
880
CHANNEL NUMBER

FIGURE 6.7

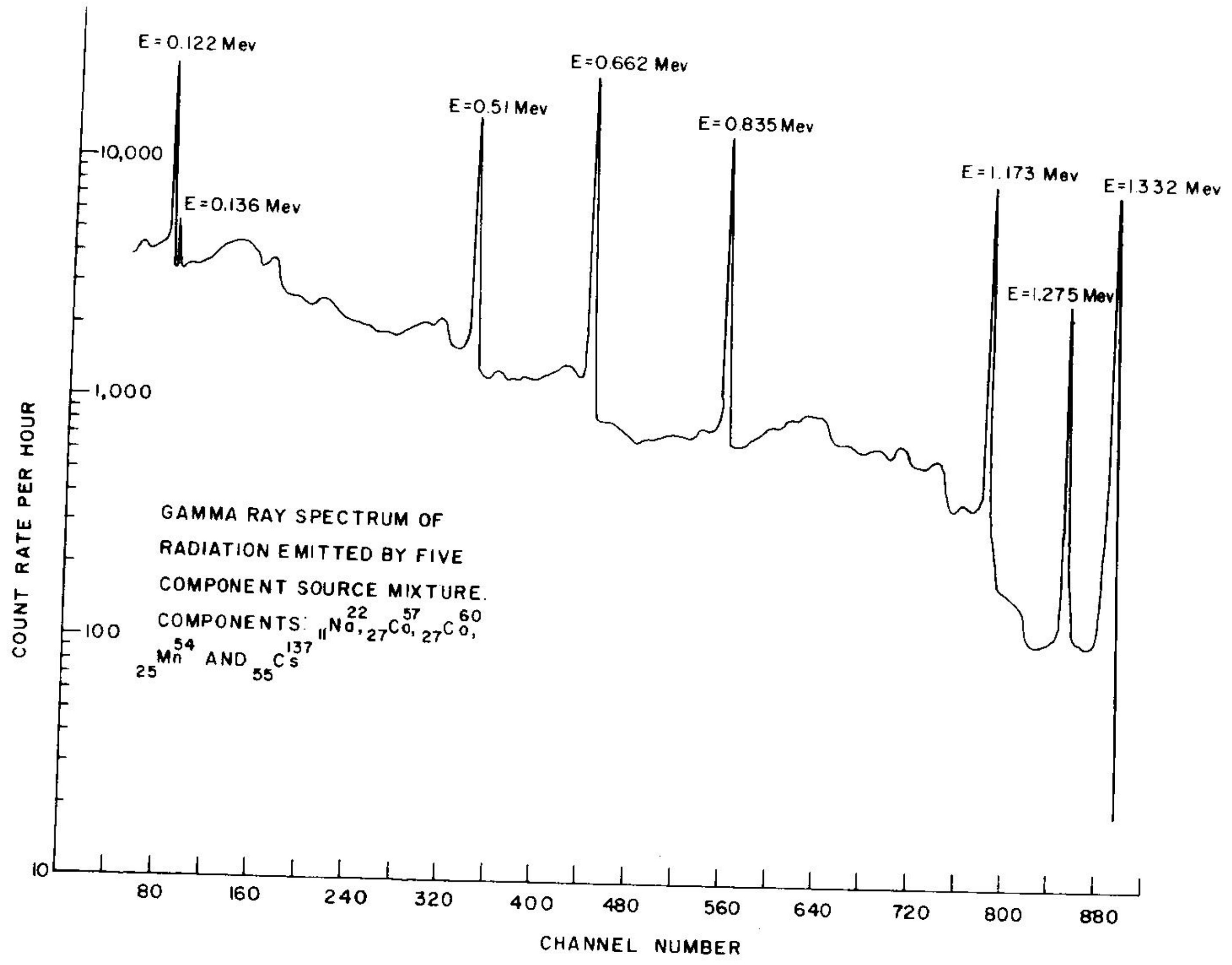

E = 0.122 Mev
E = 0.136 Mev
E = 0.51 Mev
E = 0.662 Mev
E = 0.835 Mev
E = 1.173 Mev
E = 1.275 Mev
E = 1.332 Mev
GAMMA RAY SPECTRUM OF
RADIATION EMITTED BY FIVE
COMPONENT SOURCE MIXTURE.
COMPONENTS: $_{11}Na^{22}$, $_{27}Co^{57}$, $_{27}Co^{60}$,
$_{25}Mn^{54}$ AND $_{55}Cs^{137}$
10,000
1,000
100
10
COUNT RATE PER HOUR
80
160
240
320
400
480
560
640
720
800
880
CHANNEL NUMBER

FIGURE 6.8

Future plans include both increasing the accuracy and extending of the scope of applicability of the PRGA-code. The accuracy is hoped to be increased by

a) Establishing a library tape on which the spectra of individual standards, obtained with very good counting statistics, will be recorded. Whenever data on an additional standard spectrum will become available, it will be entered into the library tape using an appropriate format.

b) A baseline and gainshift routine will be added to the existing code. According to Parr and Lucas (31) even a gainshift of one channel may change results considerably.

The scope of applicablity of PRGA is tought to be extended beyond the present one by

(a) Including a routine based on cross correlation techniques for locating peaks.

(b) Including an option for locating peaks emitted by the same nuclide and assigning them the same intensity.

(c) Applying nonlinear least square fit techniques i.e. to fit experimental points to the sum of a Gaussian and a linear function.

The ultimate objective in computer aided spectral decomposition as indicated by several authors (28, 30) is to have a set of activation analysis data fed to the computer and have the computer to determine the composition of the un-irradiated mixture with no further man-machine interference. Better even have the computer continually monitor the output of the spectrometer, computing and evaluating while the counting is in progress, controlling the counting time, so that the sample is counted just long enough to get the desired statistical accuracy and then request the system to change samples.

It is towards those very ambitious objectives that our future research efforts will be dedicated.

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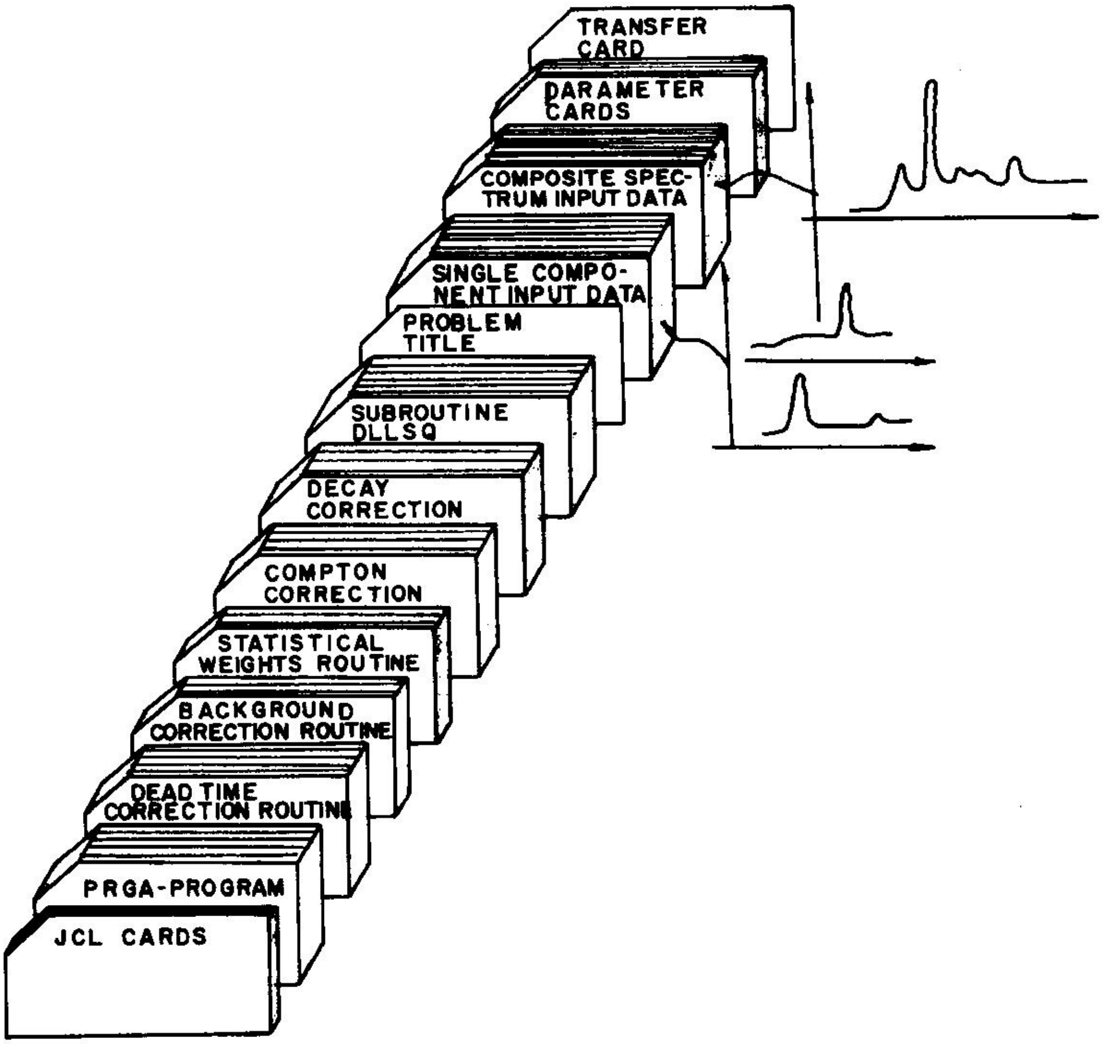
APPENDIX

ARRANGEMENT OF INPUT CARD DECK FOR PRGA-CODE AND CODE LISTING



ARRANGEMENT OF INPUT CARD DECK FOR PRGA-CODE.

FIGURE A.1

PRGA-CODE LISTING

```
C       PROGRAM....PRGA            N.KUPPUSAMY   NUCLEAR ENGG. DEPT   CAAM
C
C
        DIMENSION S(10,280),T(280),X(5),B(5),A(5,5),AUX(10),IPIV(5),BB(280
     1),KP(280),Y(280),N(280),SIG(5),DLAM(5),CC(5),YY(280)
        DOUBLE PRECISION S,T,TT,X,A,EPS,AR,D,B,AUX
C       THIS PROGRAM COMPUTES THE MIXING FACTORES,C(I),BY LEAST SQUARE FIT,
C       IF SPECTRA OF THE COMPONENTS ARE GIVEN AS INPUTS
C       SS(I,K) IS THE K-TH ORDINATE OF THE I-TH SOURCE
      1 READ 100,II,KK
    100 FORMAT(2I4)
        PRINT 111,II,KK
    111 FORMAT('1 THE NUMBER OF COMPONENTS ='I4//'0 THE NUMBER OF POINTS
      1 ='I4////)
C       II IS THE NUMBER OF SPECTRA CONSIDERED
C       KK IS THE NUMBER OF POINTS PER SPECTRUM
        READ(2,200)((S(I,K),I=1,II),K=1,KK)
    200 FORMAT(5F8.2)
        WRITE(3,222)
    222 FORMAT('1 S(I,K)-VALUES'//)
        WRITE(3,223)((S(I,K),I=1,II),K=1,KK)
    223 FORMAT ( 1H0,5F8.2)
        READ(2,300)(T(K),K=1,KK)
    300 FORMAT(2F8.2)
        WRITE(3,333)
    333 FORMAT ('1 T(K)-VALUES'////)
        WRITE(3,334)(T(K),K=1,KK)
    334 FORMAT (1H0,2F12.2)
C       BACKGROUND AND DEADTIME CORRECTIONS OF DATA
        READ(2,106)(BB(K),K=1,KK)
    106 FORMAT(7F8.2)
        READ(2,102)TAU
    102 FORMAT(F7.2)
        WRITE(3,88)
     88 FORMAT('1 VALUE OF TAU'//)
        WRITE(3,95)TAU
     95 FORMAT(1H0,F7.2)
        DO 103 I=1,II
        DO 103 K=1,KK
        S(I,K)=S(I,K)/(1-(S(I,K)*TAU))
        S(I,K)=S(I,K)-BB(K)
    103 CONTINUE
        DO 104 K=1,KK
        T(K)=T(K)/(1-(T(K)*TAU))
        T(K)=T(K)-BB(K)
    104 CONTINUE
C       COMPTON CORRECTION
C       MC IS HALFWIDTH, KKP IS TOTAL NUMBER OF PEAKS
        READ(2,201)MC,KKP
    201 FORMAT(2I4)
        WRITE(3,89)
     89 FORMAT('1 HALF WIDTH TOTAL NUMBER OF PEAKS'//)
        WRITE(3,90)MC,KKP
```

```
   90 FORMAT(2I10)
C     KP(K) IS THE CHANNEL NUMBER OF K-TH PEAK
      READ(2,202)(KP(K),K=1,KKP)
  202 FORMAT(7I4)
      WRITE(3,91)
   91 FORMAT('1 THE CHANNEL NUMBER OF K-TH PEAK'//)
      WRITE(3,202)(KP(K),K=1,KKP)
      DO 203 IK=1,KKP
      KM1=KP(IK)-MC
      KM2=KP(IK)+MC
      DMC=2*MC
      SL=(T(KM2)-T(KM1))/DMC
      DO 204 J=KM1,KM2
      DJ=J-KM1
      Y(J)=SL*DJ+T(KM1)
      N(J)=J
      T(J)=T(J)-Y(J)
      DO 204 I=1,II
      SLI=(S(I,KM2)-S(I,KM1))/DMC
      YY(J)=SLI*DJ+S(I,KM1)
      S(I,J)=S(I,J)-YY(J)
      IF(S(I,J))836,204,204
  836 S(I,J)=0.00
  204 CONTINUE
      WRITE(3,253)(N(J),T(J),(S(I,J),I=1,II),J=KM1,KM2)
  253 FORMAT (1H0,I12,6F12.2/)
  203 CONTINUE
      DO 22 J=1,II
      DO 22 I=1,II
      AR=0.0
      D=0.0
      DO 310 IK=1,KKP
      KM1=KP(IK)-MC
      KM2=KP(IK)+MC
      DO 310 K=KM1,KM2
      AR=AR+T(K)*T(K)*S(J,K)
      D=D+S(I,K)*S(J,K)*T(K)
  310 CONTINUE
      B(J)=AR
      A(I,J)=D
   22 CONTINUE
      WRITE(3,555)
  555 FORMAT('1 A(I,K)-VALUES '///)
      WRITE(3,556)((A(I,K),I=1,II),K=1,II)
  556 FORMAT (1H0,5F20.1)
      WRITE(3,557)
  557 FORMAT ('1 B(J)-VALUES'///)
      WRITE(3,558)(B(J),J=1,II)
  558 FORMAT(1H0,5F20.2)
      READ(2,600)M,NN,L
  600 FORMAT (3I4)
      READ(2,601)EPS
  601 FORMAT(E8.2)
```

```
      WRITE(3,700)EPS
  700 FORMAT ('1 EPSILON= 'E8.2)
      CALL DLLSQ (A,B,M,NN,L,X,IPIV,EPS,IER,AUX)
      WRITE(3,445)
  445 FORMAT('1 MIXING FACTORES')
      WRITE(3,444)(X(K),K=1,II)
  444 FORMAT(1H0,5F12.2)
C     DECAY CORRECTION
      READ(2,303)(DLAM(J),J=1,II)
  303 FORMAT(5E12.2)
      READ(2,304)(SIG(J),J=1,II)
  304 FORMAT(5E12.2)
      READ(2,305)T1,T2,T3,F1
  305 FORMAT(3F8.2,E10.2)
      DO 342 J=1,4
      F=DLAM(J)*T1
      G=DLAM(J)*T2
      H=DLAM(J)*T3
      FF=EXP(-F)
      GG=EXP(F-G)
      HH=EXP(F-H)
      FA=1-FF
      DNUM=SIG(J)*F1*FA*(GG-HH)
      CC(J)=X(J)*DLAM(J)/DNUM
  342 CONTINUE
      WRITE(3,777)
  777 FORMAT('1 MIXING FACTORS WITH DECAY CORRECTIONS'/////)
      WRITE(3,778)(CC(J),J=1,II)
  778 FORMAT(1H0,5F12.2)
      CALL EXIT
      END
```

```
      SUBROUTINE DLLSQ
      DIMENSION A(1),B(1),X(1),IPIV(1),AUX(1)
      DOUBLE PRECISION A,B,X,AUX,PIV,H,SIG,BETA,TOL
      IF(M-N)30,1,1
    1 PIV=0.D0
      IEND=0
      DO 4 K=1,N
      IPIV(K)=K
      H=0.D0
      IST=IEND+1
      IEND=IEND+M
      DO 2 I=IST,IEND
    2 H=H+A(I)*A(I)
      AUX(K)=H
      IF(H-PIV)4,4,3
    3 PIV=H
      KPIV=K
    4 CONTINUE
      IF(PIV)31,31,5
    5 SIG=DSQRT(PIV)
      TOL=SIG*ABS(EPS)
      LM=L*M
      IST=-M
      DO 21 K=1,N
      IST=IST+M+1
      IEND=IST+M-K
      I=KPIV-K
      IF(I)8,8,6
    6 H=AUX(K)
      AUX(K)=AUX(KPIV)
      AUX(KPIV)=H
      ID=I*M
      DO 7 I=IST,IEND
      J=I+ID
        H=A(I)
      A(I)=A(J)
    7 A(J)=H
    8 IF(K-1)11,11,9
    9 SIG=0.D0
      DO 10 I=IST,IEND
   10 SIG=SIG+A(I)*A(I)
         SIG=DSQRT(SIG)
         IF(SIG-TOL)32,32,11
   11 H=A(IST)
      IF(H)12,13,13
   12 SIG=-SIG
   13 IPIV(KPIV)=IPIV(K)
      IPIV(K)=KPIV
      BETA=H+SIG
      A(IST)=BETA
      BETA=1.D0/(SIG*BETA)
      J=N+K
      AUX(J)=-SIG
```

```
      IF(K-N)14,19,19
   14 PIV=0.D0
      ID=0
      JST=K+1
      KPIV=JST
      DO 18 J=JST,N
      ID=ID+M
      H=0.D0
      DO 15 I=IST,IEND
        II=I+ID
   15 H=H+A(I)*A(II)
      H=BETA*H
      DO 16 I=IST,IEND
      II=I+ID
   16 A(II)=A(II)-A(I)*H
      II=IST+ID
      H=AUX(J)-A(II)*A(II)
      AUX(J)=H
      IF(H-PIV)18,18,17
   17 PIV=H
      KPIV=J
   18 CONTINUE
   19 DO 21 J=K,LM,M
      H=0.D0
      IEND=J+M-K
      II=IST
      DO 20 I=J,IEND
      H=H+A(II)*B(I)
   20 II=II+1
      H=BETA*H
      II=IST
      DO 21 I=J,IEND
      B(I)=B(I)-A(II)*H
   21 II=II+1
      IER=0
      I=N
      LN=L*N
      PIV=1.D0/AUX(2*N)
      DO 22 K=N,LN,N
      X(K)=PIV*B(I)
   22 I=I+M
      IF(N-1)26,26,23
   23 JST=(N-1)*M+N
      DO 25 J=2,N
      JST=JST-M-1
      K=N+N+1-J
      PIV=1.D0/AUX(K)
      KST=K-N
      ID=IPIV(KST)-KST
      IST=2-J
      DO 25 K=1,L
      H=B(KST)
      IST=IST+N
```

```
      IEND=IST+J-2
      II=JST
      DO 24 I=IST,IEND
      II=II+M
   24 H=H-A(II)*X(I)
      I=IST-1
      II=I+ID
      X(I)=X(II)
      X(II)=PIV*H
   25 KST=KST+M
   26 IST=N+1
      IEND=0
      DO 29 J=1,L
      IEND=IEND+M
      H=0.D0
      IF(M-N)29,29,27
   27 DO 28 I=IST,IEND
   28 H=H+B(I)*B(I)
      IST=IST+M
   29 AUX(J)=H
      RETURN
   30 IER=-2
      RETURN
   31 IER=-1
      RETURN
   32 IER=K-1
      RETURN
      END
```

-Leak Test for Liquid Source-

HEALTH PHYSICS ASSAY REPORT

Bldg. ______ Room ______
Date 7/6/72
Taken by Vallecillo
Counted by "
Date counted 7/6/72

☐ Notify Dr. Gileadi on ext. ______ of results. ☐ Notified.
☐ Send copy of report to ______ Code ______

To be counted for
☐ ALPHA
Background ______ c/m Geometry ______
☑ BETA-GAMMA
Background 29 c/m Geometry 2π

Sample Number	Beta-Gamma	Alpha
1	28	
2	27	
3	38	
4	28	
5	34	
6	25	
7	32	
8	33	
9	33	
10	28	
11	24	
12	79	
13	39	
14	33	

Sample Number	Beta-Gamma	Alpha
15	28	
16	28	
17	34	
18	39	
19	32	
20	29	
21	37	
22	27	
23	25	
24	41	
25	28	
12	82	

Identification of Samples:

Remarks: