Technical Report No. 32-373

Least-Square Analysis of Gamma-Ray Pulse Height Spectra

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PREFACE

This paper represents one phase of research in partial fulfillment of requirements of doctorate degree supported by the Michigan Memorial Phoenix Project and the Atomic Energy Commission. The research is now continuing under sponsorship of the National Aeronautics and Space Administration Contract NAS 7-100.

ABSTRACT

In this analysis the pulse height spectrum due to a polyenergetic distribution of gamma rays is synthesized by using a series of normalized pulse height distributions resulting from the monoenergetic components in the incident beam. All of these monoenergetic pulse height distributions are weighted so their sum is a best fit, based upon a least-square criterion, to the experimentally determined polyenergetic pulse height distribution. There is difficulty in the application of least-square technique to the analysis of pulse height spectra because the problem is nonlinear in energy. In the technique described here, this difficulty has been overcome by using linear methods of solution, but applying the constraint that only positive or zero values be allowed for the intensities or amplitudes of the various monoenergetic components.

The method of solution proceeds in the following manner. The energy or pulse height region over which the analysis is to be performed is broken up into discrete segments. A monoenergetic pulse height spectrum corresponding to the energy at the midpoint of this segment is then obtained. The width of the segment is determined by the resolution of the system. In cases where the exact position of a photopeak can be determined, a monoenergetic pulse height spectrum corresponding to this energy is included. A fit over the total polyenergetic pulse height spectrum is performed by using the least-square and assuming that the amplitudes or intensities of all but two of the monoenergetic pulse height spectra are zero. The amplitudes of these two non-zero components are then determined. If the amplitudes are positive, a third component is added and the fit is repeated. If there is a negative amplitude, the corresponding component is eliminated from the analysis, (i.e., the amplitude is set equal to zero) and the analysis is continued by adding another component. The process continues after each addition and least-square analysis. One component is added at a time, and negative components are eliminated until all the monoenergetic components determined for the energy region have been tested. This method of solution then indicates the energy components and the corresponding amplitudes present in the polyenergetic pulse height spectrum. Error calculations are performed to determine the precision of the solution.

I. INTRODUCTION

A least-square fitting technique for the analysis of complex gamma-ray pulse height spectra has been developed. In this analysis, the pulse height spectrum due to a polyenergetic distribution of gamma rays is synthesized by using a series of normalized pulse height distributions resulting from either the monoenergetic components in the incident beam or the pulse height characteristic of various possible elements in the source. All of these pulse height distributions are weighted so that their sum is a best fit, based upon a least-square criterion, to the experimentally determined polyenergetic pulse height distribution. There is difficulty in the application of least-square technique to the analysis of pulse height spectra because the problem is nonlinear in energy. This difficulty has been overcome by using linear methods of solution, but applying the constraint that only positive or zero values be allowed for the intensities or amplitudes of the various monoenergetic components.

II. APPLICATION OF THE PRINCIPLE OF THE LEAST-SQUARE

A. Formulation

When a number of gamma rays are incident upon a NaI (Tl) crystal, the measured pulse height spectrum is made up of a summation of the photopeaks and compton continua of the various monoenergetic components. That is, if ρ_i is the total number of counts in channel i, then

$$\rho_i = \sum_n B_{in} \tag{1}$$

where B_{in} is the number of counts occurring in channel i due to the interactions of gamma ray of energy E_n with the crystal.

This can also be written as

$$\rho_i = \sum_n \beta_n A_{in} \tag{2}$$

where A_{in} is the normalized number of counts occurring in channel i due to the interaction of gamma rays of energy E_n with the crystal, and

$$\beta_n = \frac{B_{in}}{A_{in}}$$

The A_{in}'s can be obtained by using, for example, monoenergetic emitters placed in the same geometrical configuration as that of the polyenergetic emitter used to measure ρ_i.

Due to the variance in the determination of ρ_i and $\beta_n A_{in}$, $\beta_n A_{in}$ cannot be simply determined from an inversion of Eq. 2. Therefore, the most probable values of $\beta_n A_{in}$'s are determined based on the least-square criterion, that is

$$\sum_i \omega_i \left(\rho_i - \sum_n \beta_n A_{in}\right)^2 \quad \text{is to be, or approaches minimum} \tag{3}$$

where ω_i is the statistical weight and $\omega_i \sim 1/\sigma_i^2$. In the simplest case $\sigma_i^2 \sim \rho_i$, if the counting time in each chan-

nel is constant and if it is assumed that there is no variance in A_{in}. The summation is over all channels i and all energies n.

B. Method of Obtaining Minimum for Least-Square Fit

1. Incident Gamma Flux Discrete in Energy (energy distribution known and intensities required)

A number of algorithms can be used to obtain the minimum required in Eq. (3). The method of solution used depends upon how much is known about the incident flux. The simplest case is considered first. In this case the gamma ray energy distribution is known, and it is desired to determine the intensity.

The polyenergetic spectrum given by the ρ_i is measured, and since the energy distribution is known, the monoenergetic components (A_{in}) are known. The minimum is therefore obtained by taking the partial derivative with respect to β_k for each of the p monoenergetic components. Each derivative is then set equal to zero. Thus:

$$\frac{\partial M}{\partial \beta_k} = -2 \sum_i \omega_i \left(\rho_i - \sum_n \beta_i A_{in} \right) A_{ik} = 0 \qquad (4)$$

for k = 1, 2, ..., p. There are thus p linear equations to be solved for the β's. Equation (4) can be expressed in matrix notation, as follows (Ref. 1):

$$\mathbf{A}\omega\rho - (\mathbf{A}\omega A)\beta = 0 \qquad (5)$$

where β is a vector of the β_k's.

A is a p by n matrix of the pulse height spectra, n is the maximum pulse height, $\mathbf{A}$ is the transpose of A, and ω is a diagonal matrix of the ω_i's.

Solving for β, it is found that

$$\beta = (\mathbf{A}\omega A)^{-1} \mathbf{A}\omega\rho \qquad (6)$$

The calculation described in Eq. (6) has been programmed for the IBM 7090 computer and can handle up to 40 monoenergetic pulse height spectra and up to 250 values for each pulse height spectrum.

An application of this method is now considered. The emitter's energy distribution is known. The problem is to determine the relative intensities of these energy components. The emitter chosen is I^{131}. The gamma-ray energies in the spectrum of I^{131} are 0.722 Mev, 0.637 Mev, 0.364 Mev, and 0.284 Mev. Another gamma ray which is a possible contaminant is also noticed at 0.5 Mev. The measured pulse height spectrum is shown in Fig. 1. A

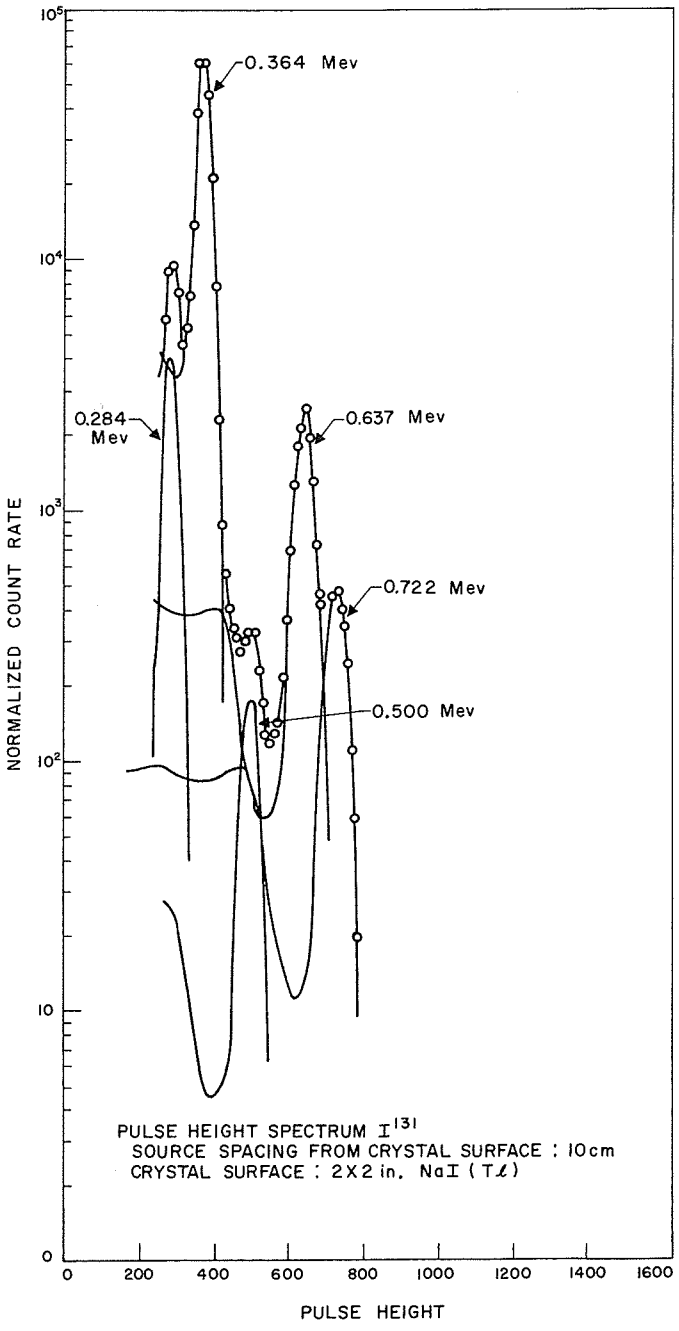

Fig. 1. Pulse height spectrum, I^{131}

point source of I^{131} was placed 10 cm from the top of a 2 x 2 in. NaI(Tl) crystal. The shapes of the monoenergetic pulse height spectra were determined by using monoenergetic sources and by extrapolating from these spectra the pulse height spectra for the energies desired. These spectra were normalized so that the area under the photopeak was unity. The curves were normalized in this manner so that the β's obtained would equal the area under the photopeak. Thus it is only necessary to

divide by the absolute peak efficiency to determine the intensity. A least-square fit is made using Eq. (6). The results are shown in tabular form in Table 1 and graphically in Fig. 1. Results for two experimentally determined decay schemes for I^{131} as determined by other investigators (Refs. 2, 3), are also presented to show the close agreement with the results obtained by using the least-square fitting technique.

Table 1. I^{131} gamma-ray spectrum

Energy, Mev	Least mean square	Other experimental results	
		Ref. 2	Ref. 3
0.284	5.2 ±0.7	6.0	4.2
0.364	100.0 ±9.0	100	100
0.500[a]	0.5 ±0.1		
0.637	10.2 ±1.2	10	7.2
0.722	2.4 ±0.3	3	2.4

[a] Possible contaminant

The errors were determined by a method described later in this Report. It was also assumed that the intrinsic efficiencies are known to within ±5%. This assumed error was determined by comparing theoretical and experimental results obtained for the efficiencies of 2 × 2 in. NaI(Tl) crystals (Refs. 4, 5, 6, 7).

2. Discrete Incident Energy Spectrum (both the energy distribution and intensity of the incident beam unknown)

The difficulty in the application of the technique lies in the method of obtaining the minimum. The minimization should be made with respect to both β_n and A_{in}; A_{in} is a function of both pulse height and energy while β_n is only a function of energy. Since the pulse height spectra (A_{in}) is not known analytically as a function of pulse height and energy, it is extremely difficult to attempt to minimize Eq. (3) with respect to the A_{in} (i.e., numerical methods would introduce large errors in the calculation). Therefore, the following method is used.

The energy spectrum under consideration is divided into discrete increments. A monoenergetic pulse height distribution corresponding to each increment is included. The energy components or increments to be chosen depend upon the photopeaks observed in the measured distribution. In those regions where the photopeaks are not obvious, the energy region is divided up depending upon the energy resolution of the system. Now, ideally one can use Eq. (6) to obtain the value β_n for the various energy components. If a given energy component m is not present, β_m should be zero or the statistical variance in β_m should be greater than β_m itself. The presence of these zeroes in the inverse transformation leads to the possibility of obtaining negative solutions, which in turn leads to oscillating components in the solution of Eq. (6). This problem is treated in detail in a work by W. R. Burrus (Ref. 8). This paper points out that the source of error in unscrambling scintillation data by the incremental technique [i.e., simple inversion of Eq. (1)] can be attributed to an error amplification when the basic equations are solved exactly. As is stated, this amplification is caused by an attempt of the exact solution to restore rapidly fluctuating components in the original gamma ray spectrum which have been attenuated below the statistical error level by the instrumental response. A first attempt to smooth out this fluctuation was made by the author using the least-square technique described above. A further smoothing can be obtained by requiring not only that Eq. (3) lead to a minimum, but that the solution for the β's be either positive or zero.

A problem demonstrating this difficulty is now considered. The problem is to determine the energies and intensities of the singlet spectrum of W^{187}. The measured pulse height spectrum is shown in Fig. 2. Certain energies are easily identified from the resolved photopeaks. In Table 2, the various energies that were assumed present are tabulated, and four iterations using the least-square fitting technique are presented.

The negative β is obtained for the 0.440 Mev gamma. The intensity of the decay scheme of W^{187} in relation to the other gamma rays present is almost zero, and is therefore lost in the background. This has been established by other experimental and theoretical calculations (Ref. 9).

An interesting result to be noted is that a variation in the choice of energy components in one region does not seem to affect the value of β in the other regions. In the 0.730-0.866 Mev region, the energies cannot be resolved as separate (i.e., there is loss of resolution due to the obliteration of the information by the detector), but the results obtained show that the number of gamma rays remains constant, although the distribution changes depending upon the components chosen to represent the region. These results seem to hold for the other cases investigated. Thus it seems that when the energy spectrum in a given region cannot be resolved, because of the finite resolution of the detection system, only the

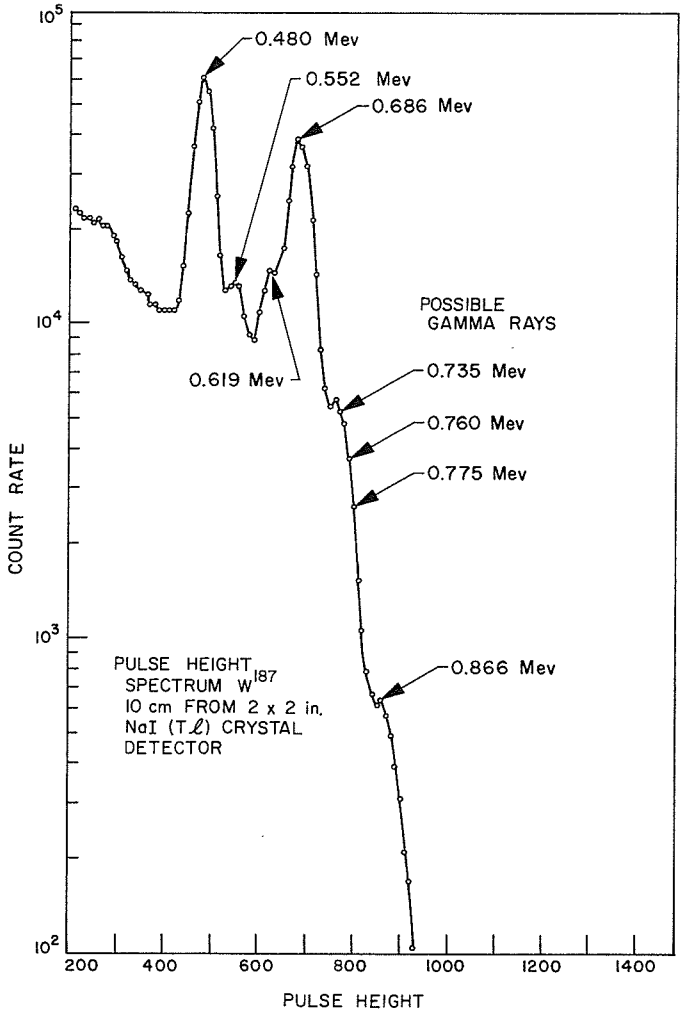

Fig. 2. Pulse height spectrum, W^{187}

total number of gamma rays in the region can be determined. The exact energy distribution in that region cannot be determined. The ability to resolve two energies can be related to the width of the photopeak at half maximum. This problem has been considered theoretically by Burrus (Ref. 8). There seems to be a minimum separation in the choice of monoenergetic components to be used in a given region, depending upon the width of the photopeak at half maximum. This choice will also depend greatly upon how well numerically (i.e., to how many significant figures) the monoenergetic pulse height spectra are known or can be known, since the least-square analysis depends upon the difference in these numerical values.

The results obtained in this calculation for W^{187} agree substantially with other experimental and theoretical calculations, and are discussed further in a paper by R. G. Arns and M. L. Wiedenbeck (Ref. 9). In this paper, the decay scheme of W^{187} is discussed in detail.

A more general method of solution for the unfolding of gamma-ray pulse height spectra has been developed. Before the method is outlined, a simple problem is presented to help clarify the discussion.

A measurement is made with a three-channel pulse height analyzer. The following data is obtained:

Channel i	*Counts*
1	3
2	2
3	3

It is known that the measured spectrum is some linear combination of the following functions:

Table 2. Results of least-square analysis of W^{187} spectrum

Energy, Mev	for Data 731	for Data 741	for Data 733	for Data 743
0.866	0.464 ±0.008	0.462 ±0.008	0.438 ±0.007	0.437 ±0.007
0.775	1.93 ±0.06	1.94 ±0.06	2.95 ±0.02	2.94 ±0.04
0.760	1.72 ±0.09	1.69 ±0.09		
0.735	0.0245 ±0.0615	0.0383 ±0.0615	1.11 ±0.03	1.11 ±0.03
0.686	22.7 ±0.06	22.5 ±0.06	21.8 ±0.05	21.7 ±0.05
0.619	7.36 ±0.03	7.33 ±0.03	7.42 ±0.03	7.40 ±0.03
0.552	6.02 ±0.03	6.02 ±0.03	6.17 ±0.03	6.18 ±0.03
0.480	25.9 ±0.06	25.6 ±0.06	26.1 ±0.06	26.0 ±0.06
0.440	−0.601 ±0.035		−0.223 ±0.036	
0.301	0.112 ±0.032	0.136 ±0.032	0.115 ±0.032	0.123 ±0.032
0.256	0.867 ±0.040	0.850 ±0.040	0.806 ±0.040	0.799 ±0.040

Channel i	NORMALIZED COUNT Ai_1	Ai_2	Ai_3
1	1	1	1
2	1	1	0
3	1	0	0

The following matrices are formed according to previous definitions:

$$A = \begin{pmatrix} 1 & 1 & 1 \\ 1 & 1 & 0 \\ 1 & 0 & 0 \end{pmatrix} \tag{7}$$

$$\mathbf{A} = \begin{pmatrix} 1 & 1 & 1 \\ 1 & 1 & 0 \\ 1 & 0 & 0 \end{pmatrix} \tag{8}$$

$$\omega = \begin{pmatrix} 1/3 & 0 & 0 \\ 0 & 1/2 & 0 \\ 0 & 0 & 1/3 \end{pmatrix} \tag{9}$$

$$\rho = \begin{pmatrix} 3 \\ 2 \\ 3 \end{pmatrix}$$

Then

$$(\mathbf{A}\omega A) = \begin{pmatrix} 7/6 & 5/6 & 1/3 \\ 5/6 & 5/6 & 1/3 \\ 1/3 & 1/3 & 1/3 \end{pmatrix} \tag{10}$$

$$(\mathbf{A}\omega A)^{-1} = \begin{pmatrix} 3 & -3 & 0 \\ -3 & 5 & -2 \\ 0 & -2 & 5 \end{pmatrix} \tag{11}$$

$$(\mathbf{A}\omega A)^{-1}\mathbf{A}\omega = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 1 & -1 \\ 1 & -1 & 0 \end{pmatrix} \tag{12}$$

The β's or intensities of each component vector can be determined from Eq. (6)

$$\beta = (\mathbf{A}\omega A)^{-1}\mathbf{A}\omega\rho = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 1 & -1 \\ 1 & -1 & 0 \end{pmatrix} \begin{pmatrix} 3 \\ 2 \\ 3 \end{pmatrix} = \begin{pmatrix} 3 \\ -1 \\ 1 \end{pmatrix} \tag{13}$$

That is, the sum of the vectors (1,1,1), (1,1,0) and (1,0,0) which yields the best fit to the experimental data using the least-square criterion is 3(1,1,1) − 1(1,1,0) + (1,0,0) = (3,2,3). The residual is zero.

If it is known that the intensities must be positive, the least-square fit must be made so as to require Eq. (3) to be a minimum with the constraint $\beta_1 \geq 0$, $\beta_2 \geq 0$, and $\beta_3 \geq 0$. One could proceed by eliminating the negative element and then re-evaluating Eq. (6). The following matrices are now formed:

$$A = \begin{pmatrix} 1 & 1 \\ 1 & 0 \\ 1 & 0 \end{pmatrix} \tag{14}$$

$$\mathbf{A} = \begin{pmatrix} 1 & 1 & 1 \\ 1 & 0 & 0 \end{pmatrix} \tag{15}$$

and ω remains the same.

Then

$$(\mathbf{A}\omega A) = \begin{pmatrix} 7/6 & 1/3 \\ 1/3 & 1/3 \end{pmatrix} \tag{16}$$

$$(A\omega A)^{-1} = \begin{pmatrix} 6/5 & -6/5 \\ -6/5 & 21/5 \end{pmatrix} \tag{17}$$

$$(\mathbf{A}\omega A)^{-1}\mathbf{A}_1^{\omega} = \begin{pmatrix} 0 & 3/5 & 2/5 \\ 1 & -3/5 & -2/5 \end{pmatrix} \tag{18}$$

Using these matrices, β is found to be

$$\beta = (\mathbf{A}\omega A)^{-1}\mathbf{A}\omega\rho = \begin{pmatrix} 0 & 3/5 & 2/5 \\ 1 & -3/5 & -2/5 \end{pmatrix} \begin{pmatrix} 3 \\ 2 \\ 3 \end{pmatrix}$$

$$= \begin{pmatrix} 12/5 \\ 3/5 \end{pmatrix} \tag{19}$$

These results indicate that the best fit requiring only a positive or zero β is

$$2.4(1,1,1) + 0(1,1,0) + .6(1,0,0) = 3,2.4,2.4$$

The method just completed is rather simple, but care must be taken in eliminating the negative components. A method for minimizing quadratics subject to various constraints is described by Beale (Ref. 10). The following is an application to the above problem, and is a simplification of the work found in Ref. 11. The simplification is possible, since the only constraint required in applying the technique to pulse height analysis is that of a positive solution for the β's when minimizing the quadratic.

In this method, the quadratic given in Eq. (3) can be formed. For the conditions in the problem discussed above, Eq. 3 can be written as

$$M = 8 - 6\beta_1 - 4\beta_2 - 2\beta_3 + \frac{7}{6}\beta_1^2 + \frac{5}{6}\beta_2^2 + \frac{\beta_3^2}{3} + \frac{5}{3}\beta_1\beta_2 + \frac{2}{3}\beta_1\beta_3 + \frac{2}{3}\beta_2\beta_3 \tag{20}$$

The sum of the residual squared (M), is to be minimized and the constraint $\beta_1 \geq 0, \beta_2 \geq 0, \beta_3 \geq 0$ is to be applied.

A geometric solution of the problem can be considered with reference to Fig. 3. The method of solution proceeds in the following manner: Start at point 0 (Fig. 3), that is, assume the solution $\beta_1 = \beta_2 = \beta_3 = 0$. Keeping $\beta_2 = \beta_3 = 0$, increase β_1 in a positive direction. As β_1 increases along this direction, M will decrease until point A is reached. Point A is determined by taking the derivative of M with respect to β_1, and setting the derivative equal to zero with $\beta_2 = \beta_3 = 0$.

$$\frac{\partial M}{\partial \beta_1} = -6 + \frac{7}{3}\beta_1 + \frac{5}{3}\beta_2 + \frac{2}{3}\beta_3 = 2u_1 \qquad (21)$$

Then for

$$u_1 = \beta_2 = \beta_3 = 0, \beta_1 = 18/7$$

The coordinates of A are (18/7, 0, 0). The plane $u_1 = 0$ in the space described by β_1, β_2, and β_3 contains all points β_1 for which M is a minimum, given any values of β_2 and β_3.

Continuing the solution, it is found that increasing β_1 any further will only increase M. A change of basis is now made. Using Eq. (21), β_1 is found in terms of u_1, β_2, and β_3.

$$\beta_1 = \frac{6}{7}\left(u_1 + 3 - \frac{5}{6}\beta_2 - \frac{1}{3}\beta_3\right) \qquad (21a)$$

This substitution is now made in Eq. (19) and

$$M = \frac{2}{7} + \frac{2}{7}\beta_2 = \frac{2}{7}\beta_3 + \frac{6}{7}u_1^2 + \frac{5}{21}\beta_2^2 + \frac{5}{21}\beta_3^2 + \frac{4}{21}\beta_2\beta_3$$

Now, keeping $u_1 = \beta_3 = 0$, one changes β_2, attempting to decrease M; that is, β_2 is increased or decreased by moving along the line of intersection of the $u_1 = 0$ plane and the $\beta_3 = 0$ plane. This intersection is along the line $\overline{AKL}$ indicated in Fig. 3. The problem is to determine in which direction to move along $\overline{AKL}$ so that M decreases. This can be done by taking the derivative of M Eq. (20) with respect to β_2.

$$\frac{\partial M}{\partial \beta_2} = \frac{2}{7} + \frac{10}{21}\beta_2 + \frac{4}{21}\beta_3 + \frac{10}{7}u_1 \qquad (22)$$

at

$$\frac{\partial M}{\partial \beta_2} = \beta_3 = u_1 = 0, \beta_2 = -\frac{3}{5}$$

In order to decrease M, β_2 must move in a negative direction. This cannot be allowed because of the constraints; therefore β_2 must be made zero. In this case

$$\beta_2 = u_2' \qquad (23)$$

and again substitution is made in Eqs. (20) and (21). The plane $u_2' = 0$ is the plane of all values of β_2, given any β_1, β_3 for which M is a minimum within the constraints of the problem. Of course, because of Eq. (23), this means $\beta_2 = 0$ for all values of β_1 and β_3.

Starting again at point A, an attempt is made to minimize M by increasing β_3 from zero. β_3 is increased along line $\overline{AJC}$, the intersection of the $u_1 = 0$ and $u_2' = 0$ planes. This insures that β_1 and β_2 will have values which will yield a minimum value of M within the constraint $\beta_1 \geq 0$, $\beta_2 \geq 0$ for any value of β_3. Using Eqs. (20) and (23), and taking the partial derivative of M with respect to β_3, the direction of increase or decrease of β_3 and also the value of β_3 can be determined so as to minimize M.

$$\frac{\partial M}{\partial \beta_3} = -\frac{2}{7} + \frac{10}{21}\beta_3 + \frac{4}{21}u_2 + \frac{4}{7}u_1 = 2u_3 \qquad (24)$$

for $u_3 = u_2' = u_1 = 0$, $\beta_3 = 3/5$. This value of β_3 along with the value $\beta_2 = u_1 = 0$ can be substituted in Eq. (21) to determine the value of

$$\beta_1 = \frac{6}{7}\left[3 - \frac{1}{3}\frac{(3)}{5}\right] = \frac{12}{5}$$

This is point C on Fig. 3 and corresponds to the intersections of the $u_1 = 0$, $u_2' = 0$, and $u_3 = 0$ planes. Thus, the values of β_1, β_2, and β_3 required to minimize Eq. (20) within the constraints that $\beta_1 \geq 0$, $\beta_2 \geq 0$, and $\beta_3 \geq 0$, are in order 12/5, 0, and 3/5. This is the same result as that obtained in Eq. (19). If the solution is continued ignoring the constraints, the absolute minimum (3, -1, 1) is obtained. This is point H, the intersection of the $u_1 = 0$, $u_2 = 0$, and $u_3 =$ planes. The solution will be independent of the path taken to reach the solution.

The method applied to a case where there are n values of β_n to be determined, can be outlined as follows:

1. Using Eq. (3), form the quadratic $M = M(\beta_1, \beta_2 \ldots, \beta_n)$
2. Take the partial derivative of M with respect to β_1, and let $2u_1 = \partial M/\partial \beta_1$

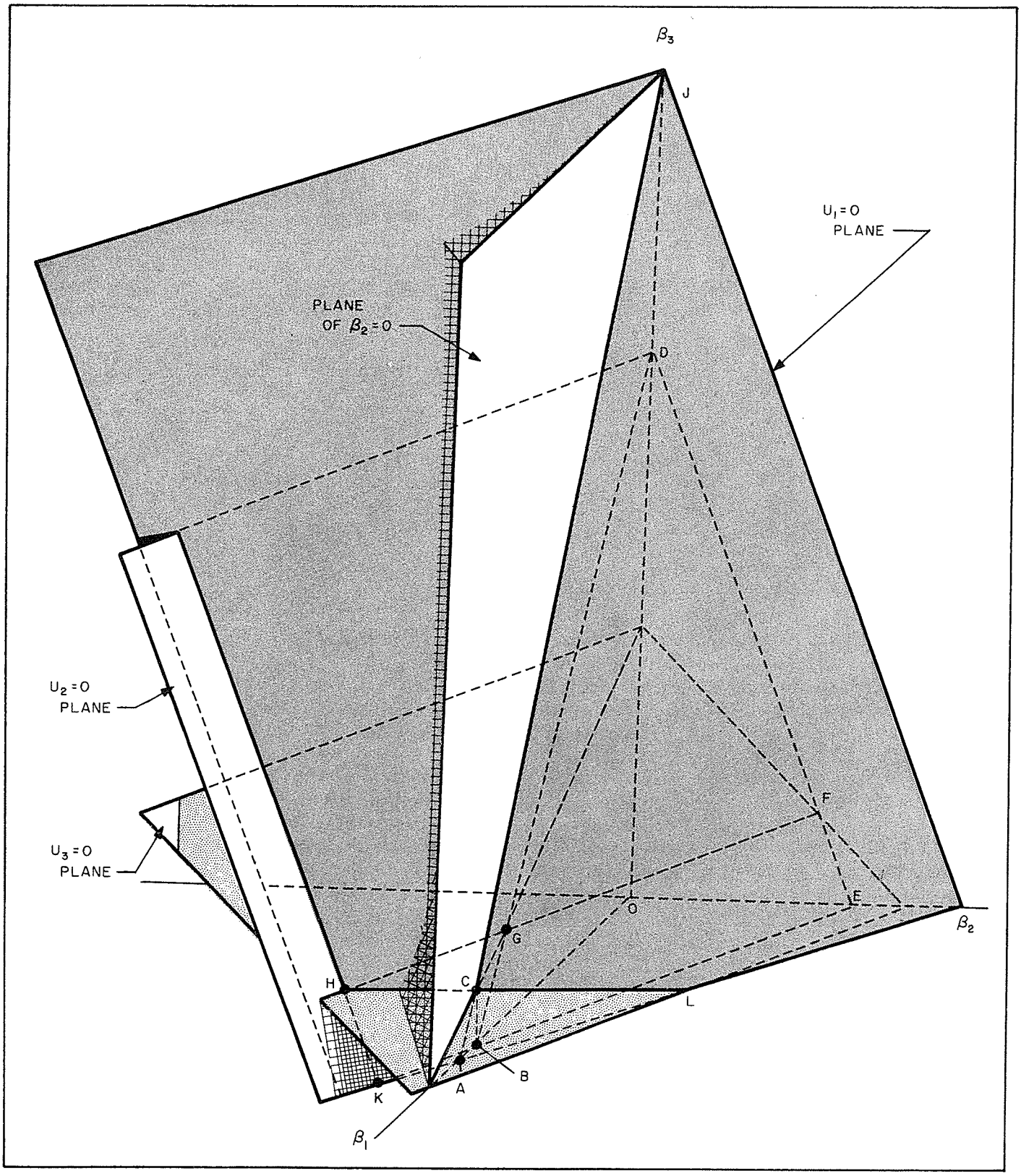

Fig. 3. Geometric solution of least-square fitting problem

3. Let $u_1 = \beta_2 = \beta_3 = \ldots = \beta_n = 0$ and solve for β_1. If $\beta_1 > 0$, then solve for β_1 in terms of $u_1, \beta_2, \ldots, \beta_n$. Substitute this value of β_1 into the equation for M. Now $M = M(u_1, \beta_2 \ldots, \beta_n)$. The first β_1 chosen will always be positive.

4. Now, using the quadratic M found in step 3, the partial derivative of M is taken with respect to β_2. Let $2u_2 = \partial M/\partial\beta_2$.

5. Let $u_1 = u_2 = \beta_3 = \ldots = \beta_n = 0$, and solve for β_2. If $\beta_2 > 0$, then solve for β_2 in terms of $u_1, u_2, \beta_3, \ldots, \beta_n$ and substitute this value in the equation for M, step 3. If $\beta_2 \leq 0$, then let $\beta_2 = u_2'$ and substitute this value of β_2 into M, step 3.

6. The above procedure is continued for all β's. At each step, the values for all the β's considered up to that point are determined. If any of these β's are negative, the variable $\beta_k = u_k'$ is changed. Further, if a previous change of variable was made which introduced a $u_j = \partial M/\partial\beta_j$, u_j must be eliminated from the function M, using the relations $u_j' = \beta_j$ and $u_j = \partial M/\partial\beta_j$ before continuing the iterative process.

7. The n values of β are found after the last iteration by setting all the u and u' vectors equal to zero.

This solution is equivalent to the following matrix approach: Assume the measured distribution is made up of only two components (e.g., the A_{i1}'s and A_{i2}'s). Least-square fitting can be used to obtain the β_1 and β_2 from Eq. (6). It has been assumed that $\beta_3 = \beta_4 = \ldots = \beta_n = 0$. If $\beta_1 \geq 0$ and $\beta_2 > 0$, add a third component (e.g., A_{i3}'s) and solve Eq. (6) for β_1, β_2, and β_3. If any of these β's are negative, that β_j is set equal to zero by eliminating the A_{ij} components from the matrix calculation involved in Eq. (6). In this manner, one of the n monoenergetic components is added at a time, and the β's for this set are determined. If any one of the β's determined in a given set is negative, that β is set equal to zero and its corresponding component is eliminated from the matrix A. The solution for the β's, after all n components have been added in the manner prescribed above, will give the values for the β's for which M is a minimum and $\beta_1 \geq 0$, $\beta_2 \geq 0, \ldots, \beta_n \geq 0$,

The number of monoenergetic pulse height spectra used in performing the above approach will depend upon the energy resolution of the system. If the energy distribution of the incident flux in some region is such that the energy separation between the various components is less than some fraction of the half-widths of the photopeaks, it may be only possible to determine the total number of gamma rays without being able to uniquely determine the energy distribution in this region. The half-width of the photopeak is a measure of the energy resoultion of the system.

The method described above has been programmed for the IBM 7090 computer. A fortran statement of this program is located in the Appendix.

At this point, a comment on the non-negativity constraint is in order. In the discussion on W^{187}, a negative intensity was obtained for a component which was almost zero in comparison to the other gamma rays in the distribution. This negative intensity may be attributed to the statistical fluctuation in the measurement of the polyenergetic spectrum, or to the uncertainty in the determination of the monoenergetic pulse height distributions. Although the negative solution due to the statistical fluctuation in the measurement of the polyenergetic spectrum might be acceptable in the analysis, it is believed that the negative introduced by the uncertainty in the monoenergetic distributions is not meaningful in the analysis because the variances in these components are not necessarily random. The non-negativity constraint is an attempt to annihilate the oscillations produced by the negative intensities.

C. Error in Calculation

Once the β's in Eq. (6) have been determined, it is possible to determine the mean square deviation in β. If it is assumed that the A_{ij}'s are known without error, this calculation is rather simple. Then, due to the variation in the measurement in ρ_i, there will be a corresponding mean square deviation in the determination of the β's (Ref. 1). Using Eqs. (4) and (6),

$$\beta_\lambda = \sum_i \sum_\nu C^{-1}_{i\lambda} A_{i\nu} \omega_i \beta_i$$

where the following definitions are used: the matrix $C = (\mathbf{A}\omega A)$ is a symmetric matrix and the elements $C_{\nu\gamma}$ of C are given by

$$C_{\nu\gamma} = \sum_i \omega_i A_{i\nu} A_{i\gamma} \qquad (25)$$

C^{-1} is the inverse of matrix of C. The elements of C^{-1} are written as $C_{\lambda\gamma}{}^{-1}$. Thus

$$CC^{-1} = \mathrm{I}$$

where I is the identity matrix with elements $\mathrm{I}_{\nu\lambda}$ and

$$\mathrm{I}_{\nu\lambda} = \sum_\gamma C_{\nu\gamma} C^{-1}_{\gamma\lambda} \qquad (26)$$

remembering that both C and C^{-1} are symmetric matrices.

Further,

$$\begin{aligned} I_{\nu\lambda} &= 1 \qquad \text{if } \nu = \lambda \\ I_{\nu\lambda} &= 0 \qquad \text{if } \nu \neq \lambda \end{aligned} \tag{27}$$

From Eq. (6), it is seen that β_λ is a linear homogeneous function of the counts under the assumption that there is no error in the A_{ij}. Thus the mean square deviation $\sigma^2(\beta_\lambda)$ corresponding to the variation in ρ_i can be written as

$$\sigma^2(\beta_\gamma) = \sum_i \sum_\nu \sum_\gamma C_{\nu\lambda}^{-1}\, C_{\gamma\lambda}^{-1}\, A_{i\nu}\, A_{i\gamma}\, \omega^2\, \sigma^2\,(\rho_i)$$

where

$$\omega_i = \frac{1}{\sigma_i^2\,(\rho_i)}$$

Then

$$\sigma^2(\beta_\lambda) = \sum_i \sum_\nu \sum_\gamma C_{\nu\lambda}^{-1}\, C_{\gamma\lambda}^{-1}\, A_{i\nu}\, A_{i\gamma}\, \omega_i$$

or

$$= \sum_\nu \sum_\gamma C_{\nu\lambda}^{-1}\, C_{\gamma\lambda}^{-1} \sum_i \omega_i\, A_{i\nu}\, A_{i\gamma}$$

Then from Eq. 25

$$\sigma^2 C(\beta_\lambda) = \sum_\nu \sum_\gamma C_{\gamma\lambda}^{-1}\, C_{\gamma\lambda}\, C_{\nu\gamma}$$

or

$$\sigma^2 C(\beta_\lambda) = \sum_\nu C_{\nu\lambda}^{-1} \sum_\gamma C_{\nu\lambda}\, C_{\gamma\lambda}^{-1}$$

and from Eqs. (26) and (27)

$$\sigma^2 C(\beta_\lambda) = C_{\lambda\lambda}^{-1} \tag{28}$$

that is, $\sigma^2(\beta_\lambda)$ can be found from the diagonal elements of the C^{-1} matrix. The mean square deviation $\sigma^2(\rho_i)$ in the simplest case is

$$\sigma^2(\rho_i) = \rho_i \tag{29}$$

Depending upon the experiment, the corresponding σ_i's can be determined and used in the above situation. The probable error can then be determined from the mean square deviation.

The above considerations are true only if it is assumed that the A_{ij}'s are known without error, that the set of A_{ij}'s chosen are the correct set, and that the set of pulse height spectra are linearly independent (i.e., there is no interference between various components). Further calculations are carried in an attempt to determine whether these problems contribute a significant error.

A factor indicating percent interference can be calculated in the following manner:

$$I_n = \frac{(C_{\gamma\lambda}^{-1})^2}{C_{\lambda\lambda}^{-1}\, C_{\gamma\gamma}^{-1}} \times 100\% \tag{30}$$

It was shown above that $C_{\lambda\lambda}{}^{-1}$ is the variance on β_λ, and is can be shown (Ref. 11) that $C\lambda\gamma^{-1}$ is the covariance of the λ'th and γ'th components. Then, Eq. (30) is a measure of interference between the λ'th and γ'th components (Refs. 11, 12).

D. The Analysis of Complex Gamma Spectra

The following experiment was designed to test the analytic method described above. A mixture of ten different elements was activated in a thermal neutron flux for a given length of time and the pulse height spectrum of the activated sample was measured as a function of time. A known amount of each element was activated in the same neutron flux for the same length of time as the mixture, and the pulse height spectrum of each of these elements was measured in the same geometrical configuration as that for the mixture. A 3 × 3 in. NaI (*Tl*) crystal with a 200 channel pulse height analyzer was used for the measurement. These pulse height spectra are shown in Figs. 4-24. The background spectrum is included as a separate pulse height distribution. Furthermore, it was noticed that due to the presence of some air in the sample, argon gas had been activated and tended to perturb the measurement of the pulse height spectra of the various other elements. A pure argon pulse height spectrum was measured and an attempt was made to subtract this effect from the spectra of the other elements. The argon spectrum was included in the analysis of the mixture spectrum. Finally, it was noticed that in the supposedly pure chromium spectrum, there was a significant amount of sodium. The contribution of the sodium spectrum to the chromium spectrum was substracted out, and a "pure" chromium spectrum was obtained.

Since the mixture and each of the various elements were activated in the same neutron flux for the same length of time, and since the standard spectra used in the analyses were those due to the activation of the various elements, the results of the least-square analysis will be in terms of the relative abundance of the element in the mixture to the amount in the standard. Because there is radioactive decay of the standard and mixture, decay time correction must be made in order to determine the difference between the absolute abundance and the relative abundance. The relative abundances obtained from the least-square analysis are tabulated in Table 3. The zeros indicate that the given element has been rejected in the fit. Interference between the various component spectra were then calculated. Those interferences which

Table 3. Relative intensities of various elements in mixture obtained using least-square analysis Mixture removed from reactor Oct. 4, 1962 at 1055

Element	Oct. 4 1122	Oct. 4 1146	Oct. 4 1259	Oct. 4 1633	Oct. 5 0827	Oct. 5 1641	Oct.6 0900	Oct. 7 1329	Oct. 11 0914
Background	9.44	9.55	33.6	3.6	4.72	4.86	5.86	6.87	1.91
Na-24	19.9	19.1	15.2	17.1	7.22	3.86	2.12	0.0	0.0
Cl-38	0.68	0.466	0.133	0.0	0.0	0.0	0.0	0.0	0.0
K-42	8.78	7.67	5.59	6.5	3.28	2.34	0.0630	0.0	0.0
Mn-56	0.0691	0.0669	0.0382	0.0166	0.00173	0.0048	0.0	0.0	0.0
Sc-46	0.0340	0.0340	0.0422	0.0442	0.0488	0.0466	0.0360	0.0431	0.0413
A-41	0.0	0.0	0.0	0.0	0.0	0.00806	0.0	0.0	0.0
As-76	10.6	12.5	11.3	9.75	6.04	4.52	3.54	1.65	0.363
Cu-64	4.45	3.49	1.94	2.91	1.77	1.38	0.183	0.148	0.0493
Cr-51	4.00	3.93	3.60	4.81	5.0	4.94	4.12	4.39	4.13
I-128	18.1	9.6	0.0	0.0	0.138	0.0274	0.0	0.0	0.132
La-140	5.68	4.79	4.41	3.98	2.30	1.91	1.57	0.989	0.199

were significantly high are plotted in Fig. 25. From these curves it can be seen that one would expect significant errors in the calculation of the backgrounds of Lanthanum, Potassium, Sodium, and Scandium. At certain times the interference may be great, but if one element decays away, the interference will become negligible.

After correcting for decay, the analysis of the mixture was obtained, and compared with the actual composition of the mixture. The results are given in Table 4. The calculated errors indicated in the table do not include interference effects.

The calculation for sodium seems to be rather insubstantial; however, it must be remembered that the sodium was noticed in the chromium standard and this may partially explain the discrepancy. Further, it should be noted that in those cases with significant interference, one element would be overestimated while the other with interference would be underestimated. The remaining results seem to be in rather good agreement within the statistical variance involved in the experiment.

The interference effect may be greatly reduced by breaking up the various standard spectra into monoenergetic components and using these components for the analysis. This group of monoenergetic spectra should be a linearly independent group.

Further, it can be seen that there is a very significant interference by background. This background should be subtracted before the analysis is performed. In the problem above, the background was very small in comparison to the counts above background, so that when the analysis was redone with the background subtracted, there were no significant differences from those reported in Table 4.

Table 4. Experimental determination of composition

Element	Prepared sample	Experiment determinations
Sodium	5.00 μg	7.7 ±1 μg
Chlorine	36.8 μg	38.0 ±3.8 μg
Potassium	0.404 mg	0.301 ±1.011 mg
Manganese	0.101 μg	0.0904 ±0.0018 μg
Scandium	14.2 μg	12.2 ±2.5 μg
Arsenic	5.00 μg	4.98 ±0.11 μg
Copper	2.23 μg	2.11 ±0.16 μg
Chromium	1.82 mg	1.56 ±0.24 mg
Iodine	2.49 μg	2.68 ±0.08 μg
Lanthonum	2.19 μg	3.02 ±.04 μg

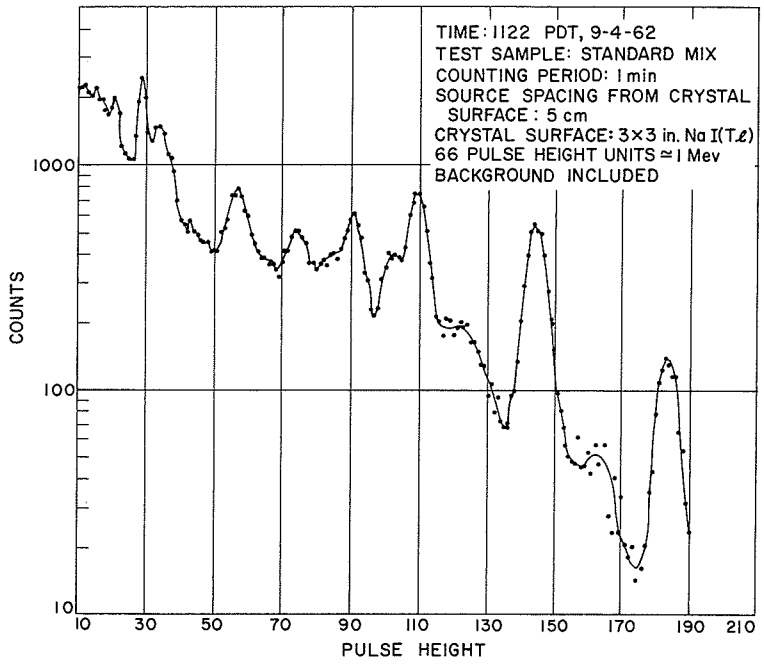

Fig. 4. Pulse height spectrum, standard mix, 1122 PDT, 9-4-62

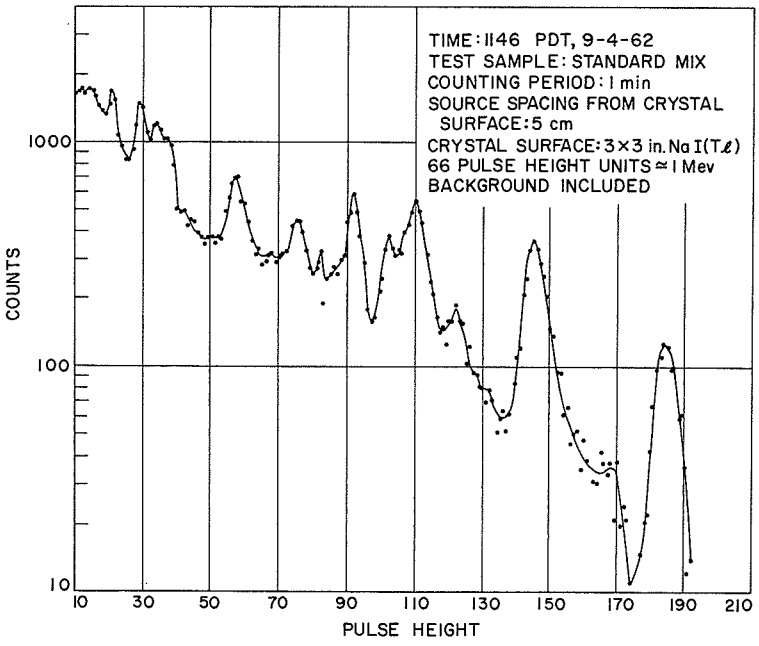

Fig. 5. Pulse height spectrum, standard mix, 1146 PDT, 9-4-62

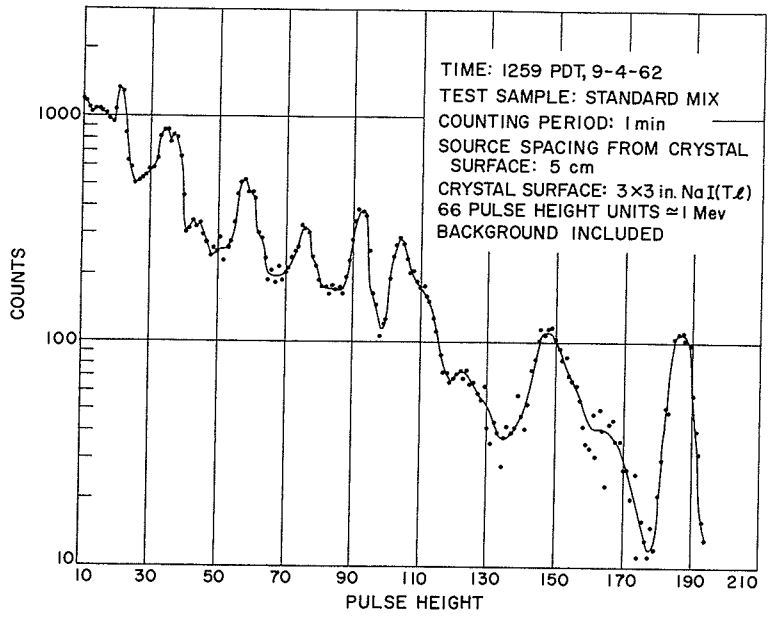

Fig. 6. Pulse height spectrum, standard mix, 1259 PDT, 9-4-62

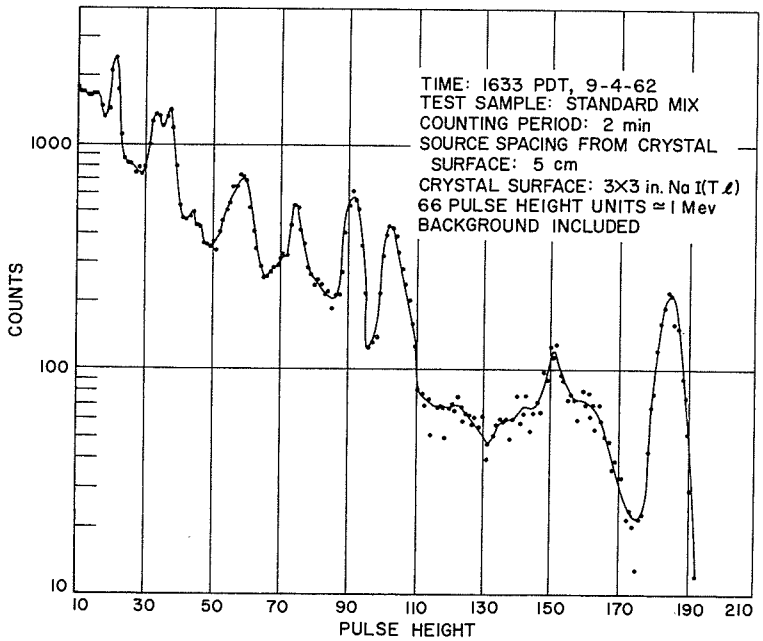

Fig. 7. Pulse height spectrum, standard mix, 1633 PDT, 9-4-62

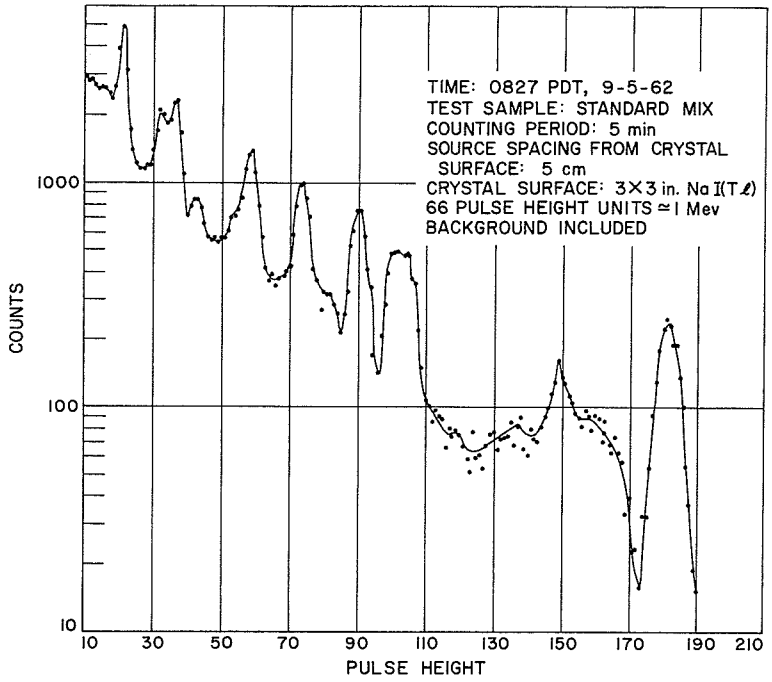

Fig. 8. Pulse height spectrum, standard mix, 0827 PDT, 9-5-62

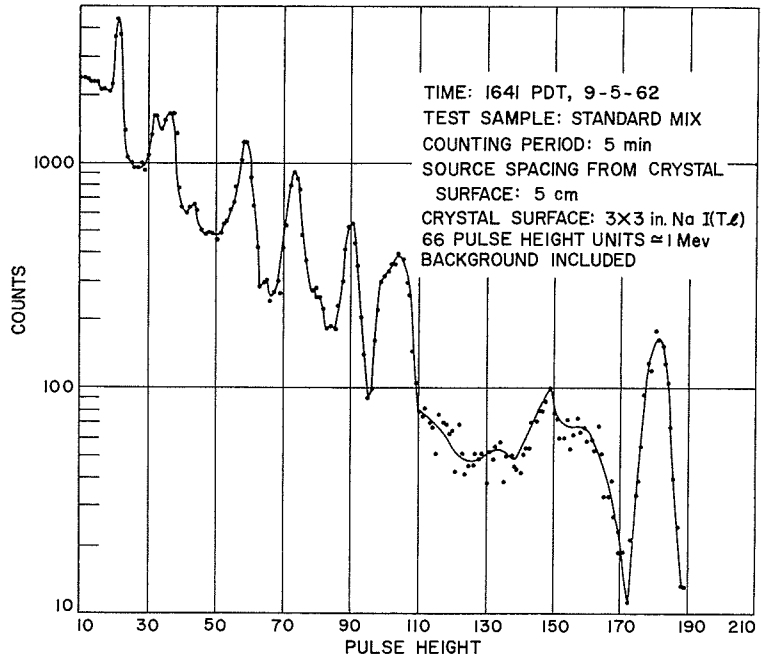

Fig. 9. Pulse height spectrum, standard mix, 1641 PDT, 9-5-62

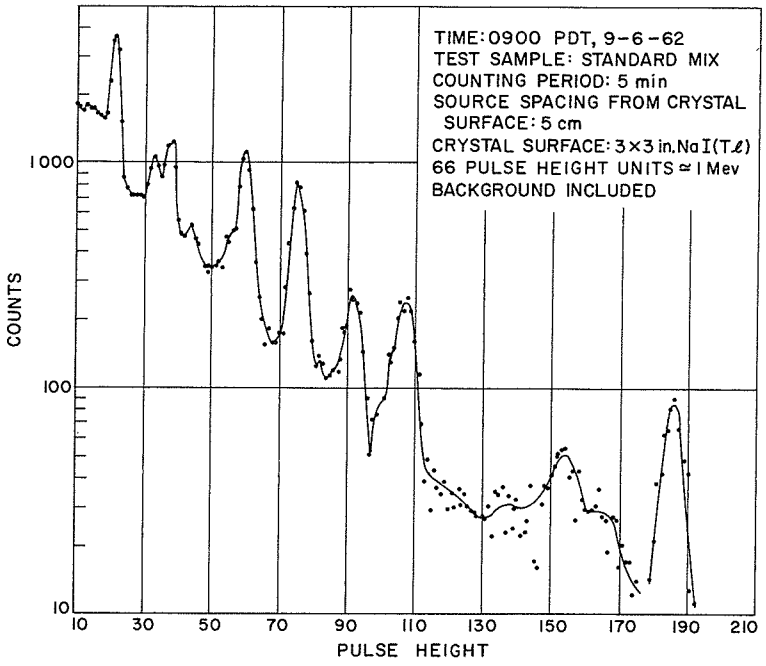

Fig. 10. Pulse height spectrum, standard mix, 0900 PDT, 9-6-62

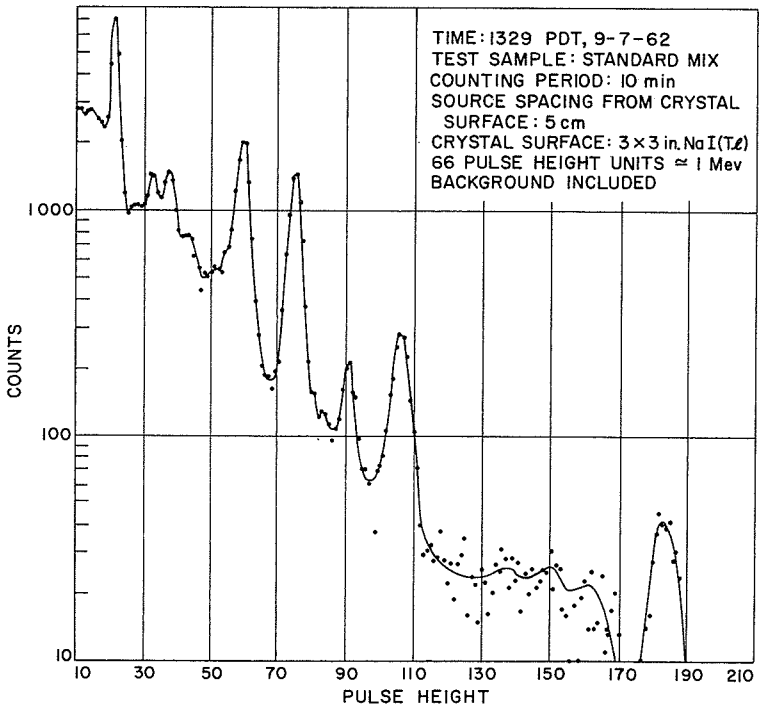

Fig. 11. Pulse height spectrum, standard mix, 1329 PDT, 9-7-62

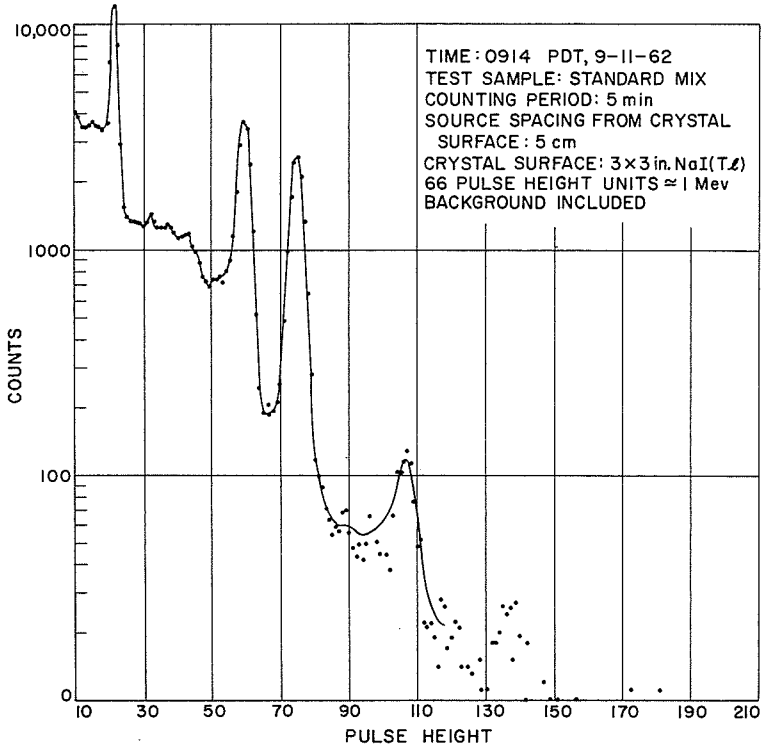

Fig. 12. Pulse height spectrum, standard mix, 0914 PDT, 9-11-62

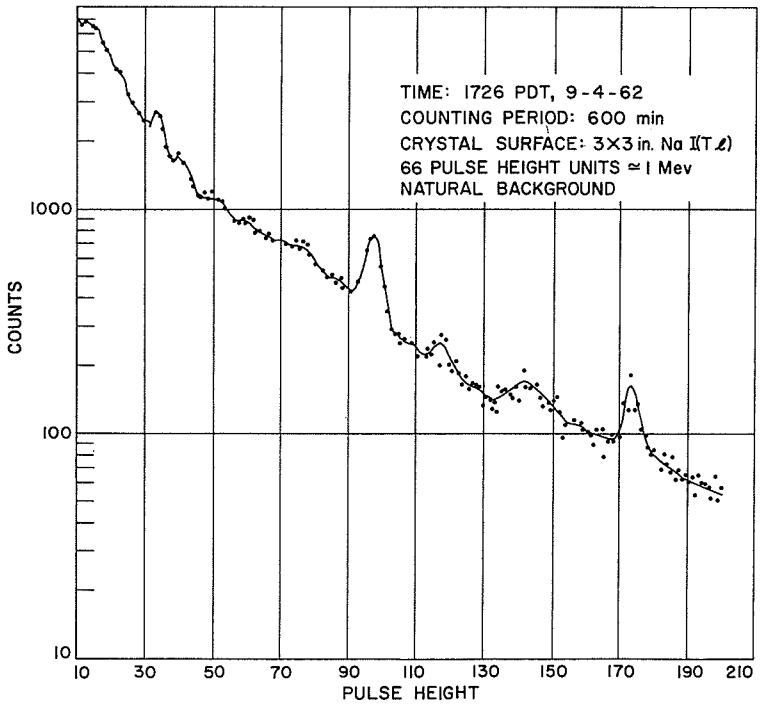

Fig. 13. Pulse height spectrum, natural background, 1726 PDT, 9-4-62

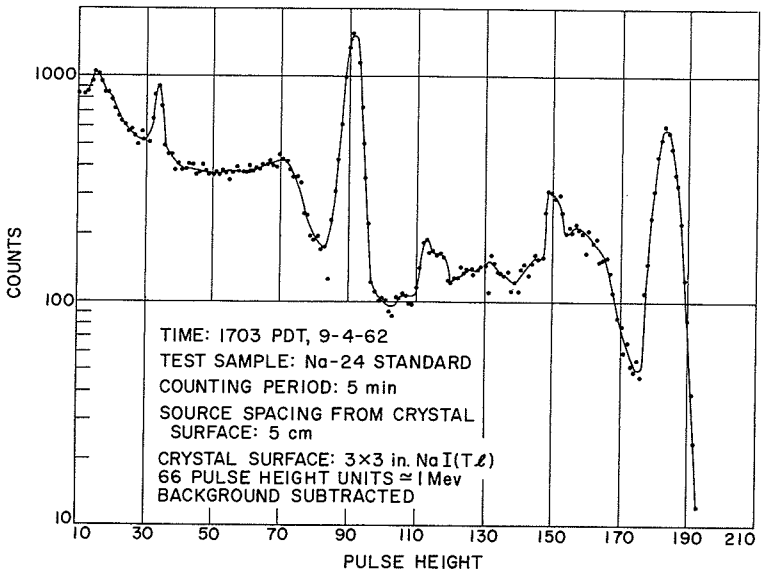

Fig. 14. Pulse height spectrum, Na-24 standard

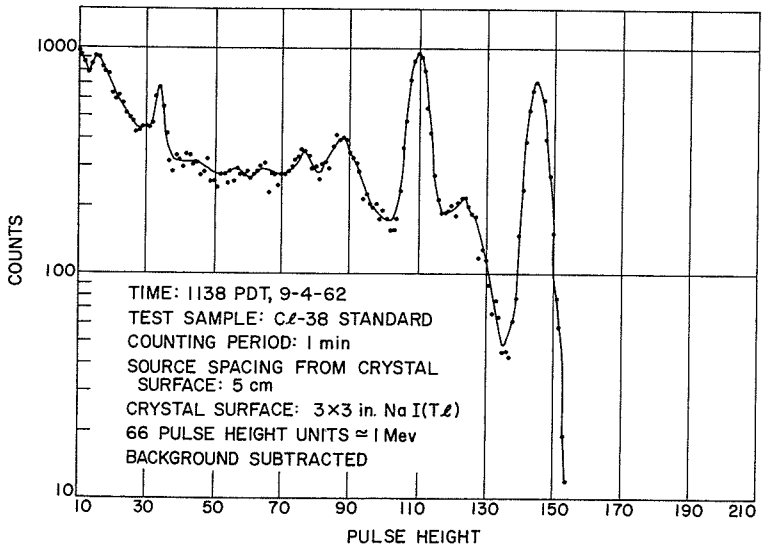

Fig. 15. Pulse height spectrum, Cl-38 standard

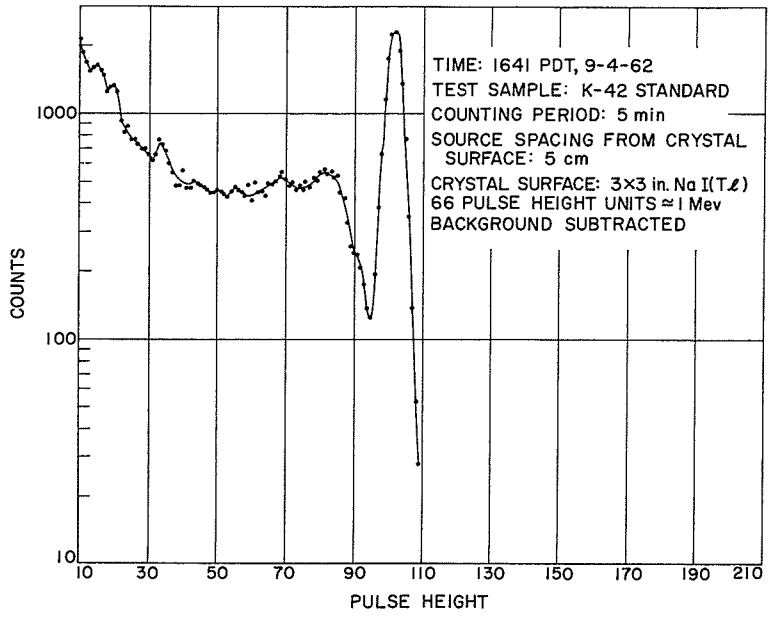

Fig. 16. Pulse height spectrum, K-42 standard

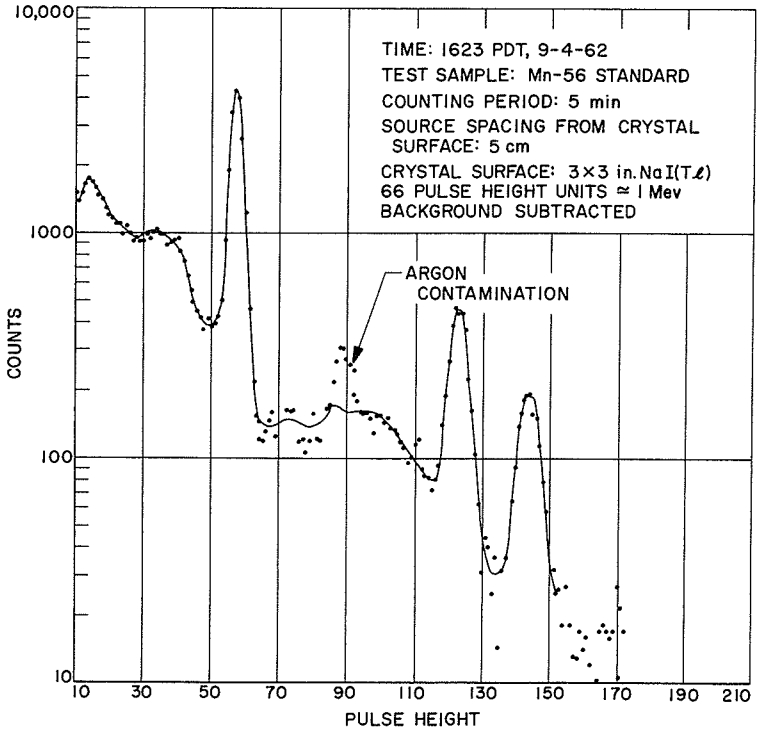

Fig. 17. Pulse height spectrum, Mn-56 standard

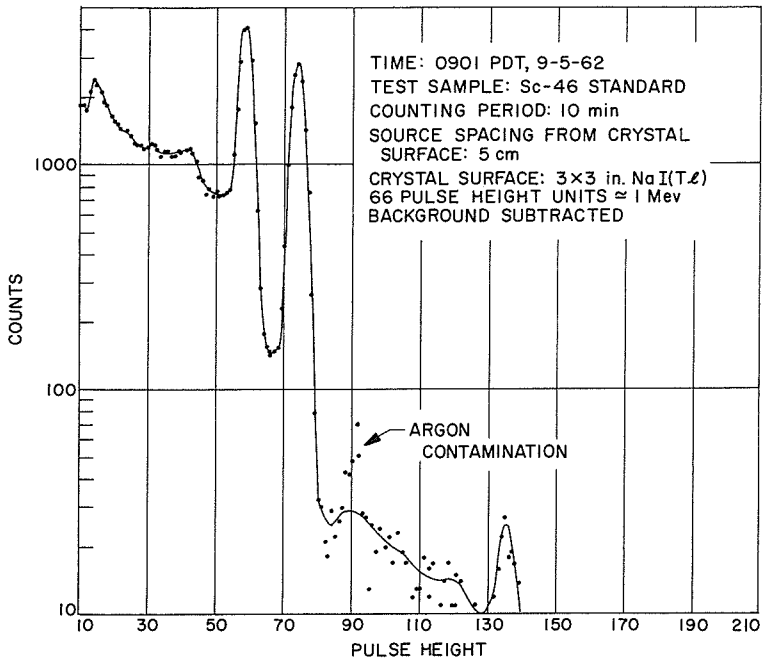

Fig. 18. Pulse height spectrum, Sc-46 standard

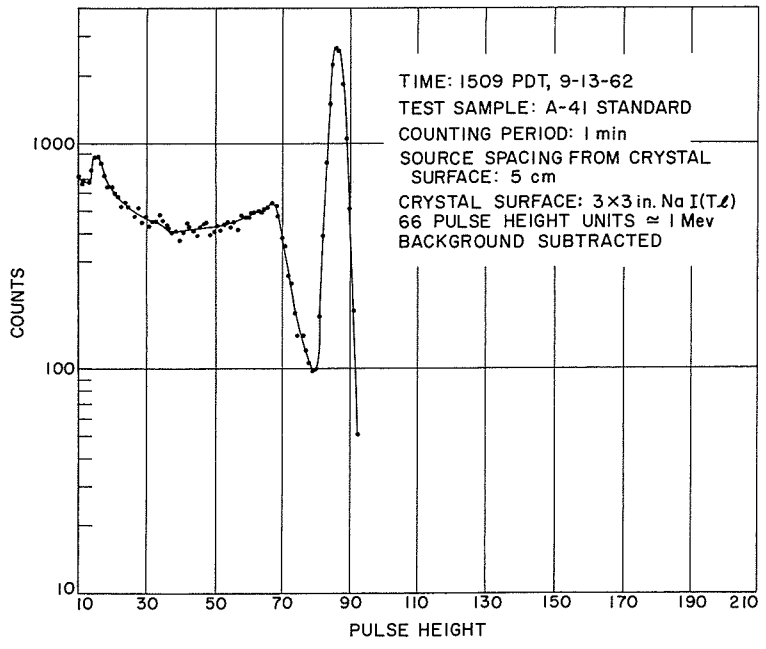

Fig. 19. Pulse height spectrum, A-41 standard

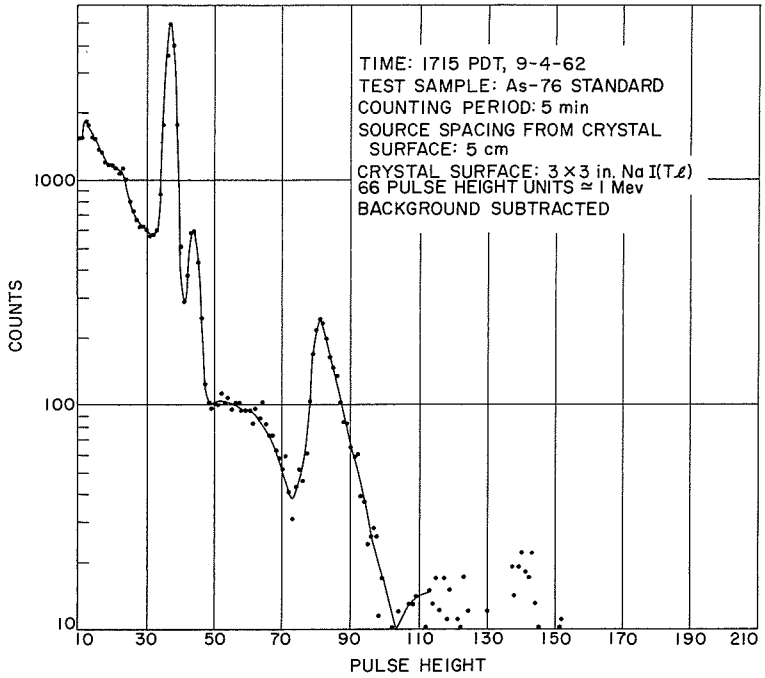

Fig. 20. Pulse height spectrum, As-76 standard

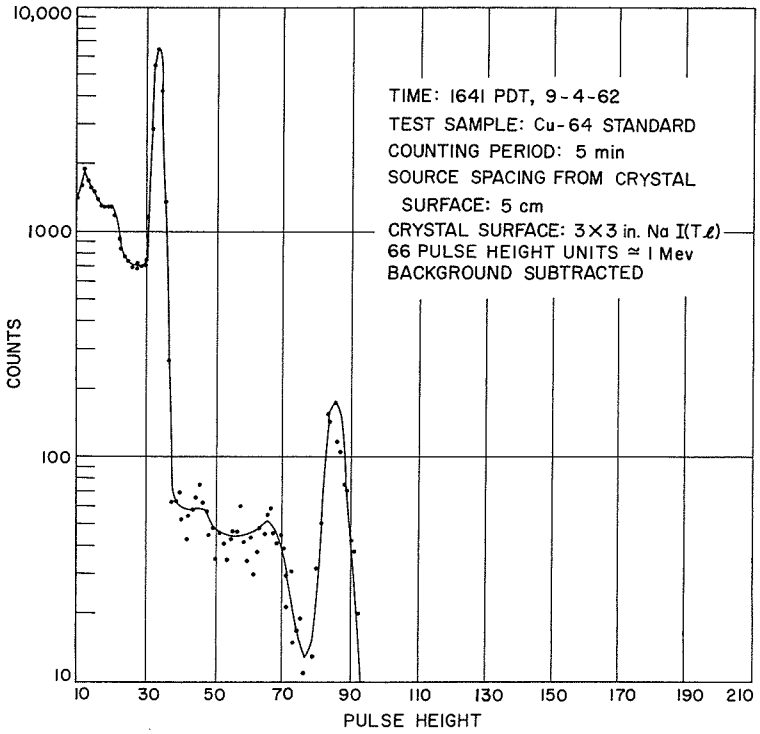

Fig. 21. Pulse height spectrum, Cu-64 standard

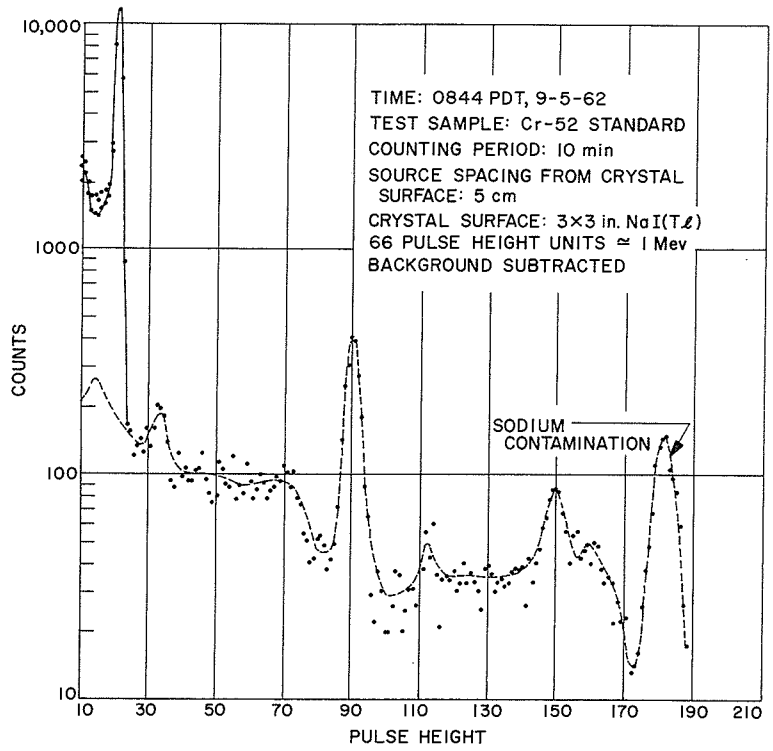

Fig. 22. Pulse height spectrum, Cr-52 standard

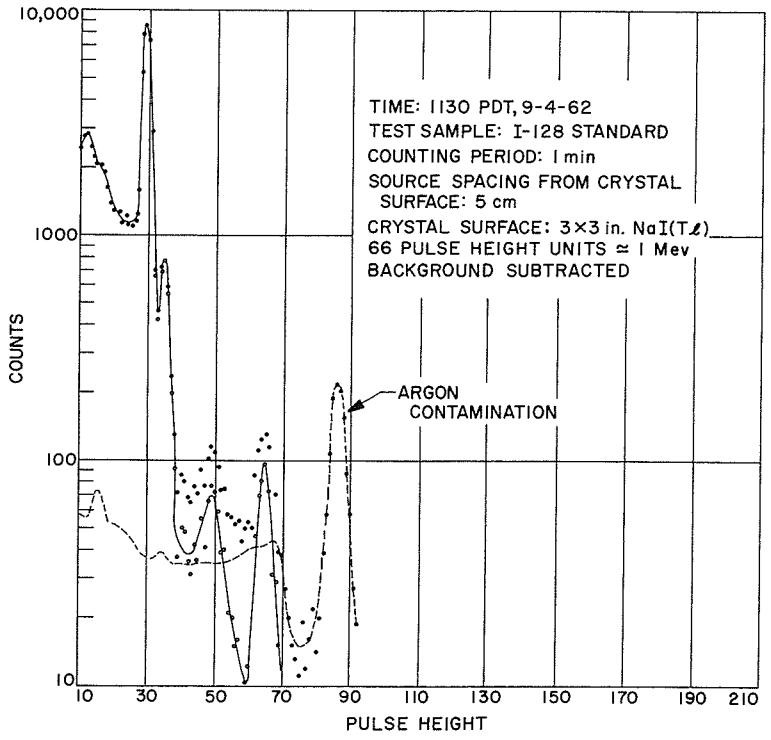

Fig. 23. Pulse height spectrum, I-128 standard

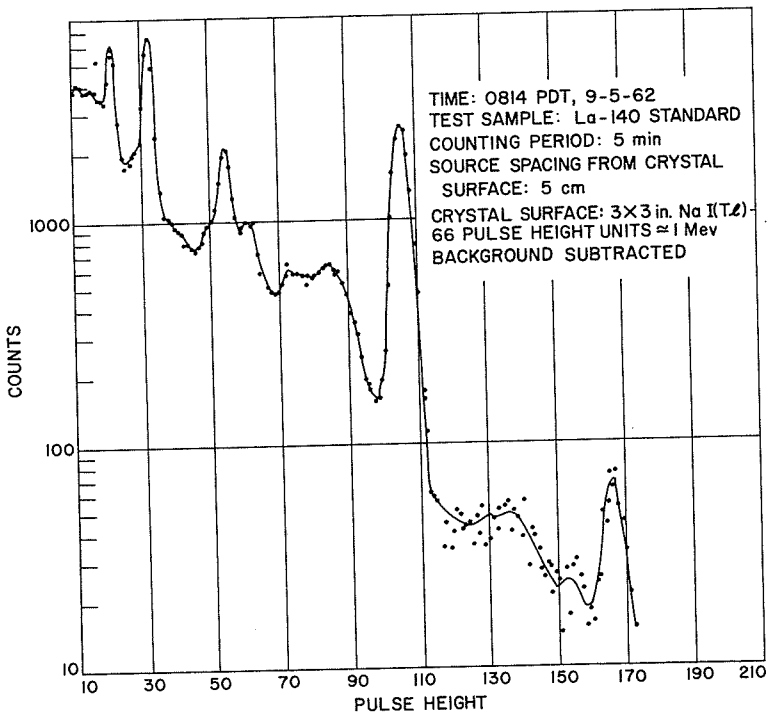

Fig. 24. Pulse height spectrum, La-140 standard

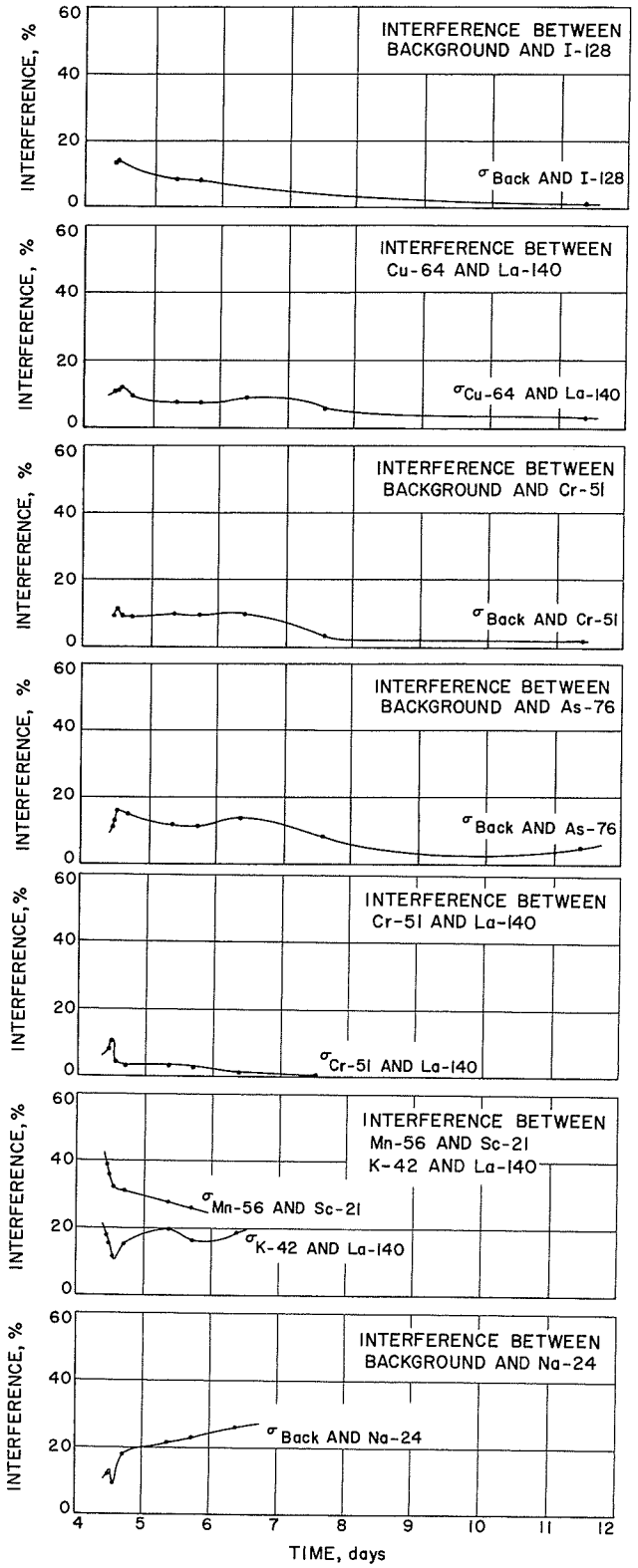

Fig. 25. Interference between various component spectra

III. CONCLUSION

Finally, it should be pointed out that the analysis is only as good as the set of fitting spectra available. Spectra such as those due to bremsstrahlung should be included in the library of functions used for analysis. At the present time, a chi-square test is being prepared for inclusion in the analysis for testing the goodness of fit. This test, together with the calculation of interference, should indicate whether such difficulties as the shifting of gain and the lack of a complete library of functions have perturbed the analysis.

ACKNOWLEDGEMENT

The author wishes to thank Professor M. Wiedenbeck, University of Michigan, for his guidance in the development of this Report; Dr. V. Guinn and his activation analysis group at General Dynamics Corporation, General Atomic Division, San Diego, California, for their aid in the preparation of samples, activation of these samples, and measurement of the pulse height distributions; and P. Poulson of the Computer Staff at JPL for the preparation of the computer code used in the analysis.

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APPENDIX

Fortran Statement, Least-Square Analysis

The following is a Fortran statement of the least-square analysis program, and was prepared by P. Poulson of the Computer Staff, Jet Propulsion Laboratory.

The size of the matrices used in the calculation is limited by the size of the memory core of the IBM 7090 at the Laboratory. The designations for the various vectors and matrices in the program are the same as those used in the formulation of the least-square analysis as presented in this Report.

A few comments with regard to the input form may aid in the understanding of the problem:

(1) There are two possible calculational MØDES;

MØDE = 0, non-iterative

MØDE = 1, iterative

The input is identical for both MØDES.

(2) NRØW is the number of rows.

(3) NCØL is the number of columns.

(4) NØM is the Omega MØDE.

NØM = −1, input the weights

NØM = 0, unity matrix equal weight

NØM = 1, set weight equal to 1/RHØ

(5) NRHØ is the identification number for complex spectrum set.

(6) NRUN is the identification number of the A matrix set.

(7) Input procedures:

a. Input A matrix by columns.

b. Input RHØ vector of size NRØW

c. Input Omega matrix if applicable (i.e., if NØM = −1).

(8) SETUP, MMULT, and MVERT are all JPL matrix package subroutines.

```
C  LEAST SQUARES ANALYSIS OF GAMMA RAY PULSE HEIGHT SPECTRA
C  BY  BUD POULSON,  JET PROPULSION LABORATORY, PASADENA, CALIFORNIA
      DIMENSION A(250,40),AT(40,250),RHO(250),T(40,40),OMEGA(250)
      DIMENSION BETA(40),LEFT(300),LIST(300),THETA(250)
      CALL SETUP(A,250,40,AT,40,250,T,40,40,RHO,OMEGA,THETA,250,1,BETA,
     X40,1)
    1 READ INPUT TAPE 5,1000,NROW,NCOL,NOM,NRUN,MODE
      IF(MODE)200,200,250
  200 DO 2 J=1,NCOL
    2 READ INPUT TAPE 5,1001,(A(I,J),I=1,NROW)
      GO TO 3
  250 FINISH=0.0
      REWIND 4
      DO 280 J=1,NCOL
      IF(J-NCOL)270,260,260
  260 FINISH=1.0
  270 READ INPUT TAPE 5,1001,(OMEGA(I),I=1,NROW)
      WRITE TAPE 4,(OMEGA(I),I=1,NROW),FINISH
  280 CONTINUE
    3 READ INPUT TAPE 5,1000,NRHO
      IF(NRHO)1,1,4
    4 READ INPUT TAPE 5,1001,(RHO(I),I=1,NROW)
      WRITE OUTPUT TAPE 6,5000
      WRITE OUTPUT TAPE 6,5006,NRUN,NRHO
      IF(NOM)5,20,10
    5 READ INPUT TAPE 5,1001,(OMEGA(I),I=1,NROW)
      GO TO 25
   10 DO 15 I=1,NROW
   15 OMEGA(I)=1.0/RHO(I)
      GO TO 25
   20 DO 22 I=1,NROW
   22 OMEGA(I)=1.0
   25 IF(MODE)26,29,26
   26 REWIND 4
      NCOL=2
      MODE=1
      READ TAPE 4,(A(I,1),I=1,NROW),FINISH
      JJ=0
      LL=1
      LEFT(1)=1
   27 READ TAPE 4,(A(I,NCOL),I=1,NROW),FINISH
      LL=LL+1
      LEFT(NCOL)=LL
   29 DO 30 J=1,NCOL
      DO 30 I=1,NROW
   30 AT(J,I)=A(I,J)*OMEGA(I)
   31 CALL MMULT(AT,A,T,NCOL,NROW,NCOL)
      IF(MODE)32,32,50
   32 K=1
      WRITE OUTPUT TAPE 6,5002
   35 DO 40 I=1,NCOL
      WRITE OUTPUT TAPE 6,5003,I
   40 WRITE OUTPUT TAPE 6,5001,(T(I,J),J=1,NCOL)
      GO TO (45,60),K
   45 K=2
   50 CALL MMULT(AT,RHO,THETA,NCOL,NROW,1)
      CALL MVERT(T,NCOL)
      IF(MODE)55,55,60
   55 WRITE OUTPUT TAPE 6,5004
      GO TO 35
   60 CALL MMULT(T,THETA,BETA,NCOL,NCOL,1)
      WRITE OUTPUT TAPE 6,5005
      WRITE OUTPUT TAPE 6,5001,(BETA(I),I=1,NCOL)
      IF(MODE)62,3,70
   62 TOTAL=0.0
      DO 67 I=1,NROW
      SUM=0.0
      DO 65 J=1,NCOL
   65 SUM=SUM+BETA(J)*A(I,J)
   67 TOTAL=TOTAL+OMEGA(I)*(RHO(I)-SUM)**2
      AROW=NROW-NCOL
      TOTAL=TOTAL/AROW
      WRITE OUTPUT TAPE 6,5007,TOTAL
      DO 69 I=1,NCOL
      IF(BETA(I))70,70,69
   69 CONTINUE
      GO TO 3
   70 KVAIL=1
      DO 100 I=1,NCOL
      IF(BETA(I))95,95,80
   80 DO 90 J=1,NROW
   90 A(J,KVAIL)=A(J,I)
      LEFT(KVAIL)=LEFT(I)
      KVAIL=KVAIL+1
      GO TO 100
   95 JJ=JJ+1
      LIST(JJ)=LEFT(I)
  100 CONTINUE
      NCOL=KVAIL
      IF(KVAIL-41)105,110,110
  105 IF(FINISH)107,107,110
  107 IF(NCOL-40)27,27,110
  110 MODE=-1
      NCOL=NCOL-1
      IF(JJ)29,29,120
  120 WRITE OUTPUT TAPE 6,5008,(LIST(I),I=1,JJ)
      GO TO 29
 1000 FORMAT(7I10)
 1001 FORMAT(7F10)
 5000 FORMAT(31H1GAMMA RAY PULSE HEIGHT SPECTRA)
 5001 FORMAT(1H0,7E16.5/(1H ,7E16.5))
 5002 FORMAT(7H0MATRIX)
 5003 FORMAT(4H0ROWI4/1H )
 5004 FORMAT(16H0MATRIX INVERTED)
 5005 FORMAT(12H0BETA VECTOR/1H )
 5006 FORMAT(4H0RUNI6,5X,7HRHO NO.I6)
 5007 FORMAT(5H0SUM=E20.8)
 5008 FORMAT(16H0COLUMNS OMITTED,10X,20I4/(26X,20I4))
      END
```

Fig. A-1. Computer fortran statement of least-square analysis