SIMPLEX OPTIMIZATION BY ADVANCE PREDICTION IN INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS.

SIMPLEX OPTIMIZATION BY ADVANCE PREDICTION IN INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS

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ABSTRACT

Though Neutron Activation Analysis is one of the most sensitive multielement analysis methods, Compton interference in complex sample matrices usually presents a problem when choosing irradiation and decay times for analysis. While various scientists have attempted to solve the problem, the aprroaches taken to date have the drawbacks of either requiring standard spectra of the sample components or not giving the optimum times automatically and simultaneously. The purpose of this thesis was to find a method to automatically and simultaneously obtain the optimum times for Instrumental Neutron Activation Analysis using the Modified Simplex Method to evaluate the best figure of merit calculated from an advance predicted spectrum of the sample.

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CHAPTER 1

1.1 OBJECTIVE OF STUDY:

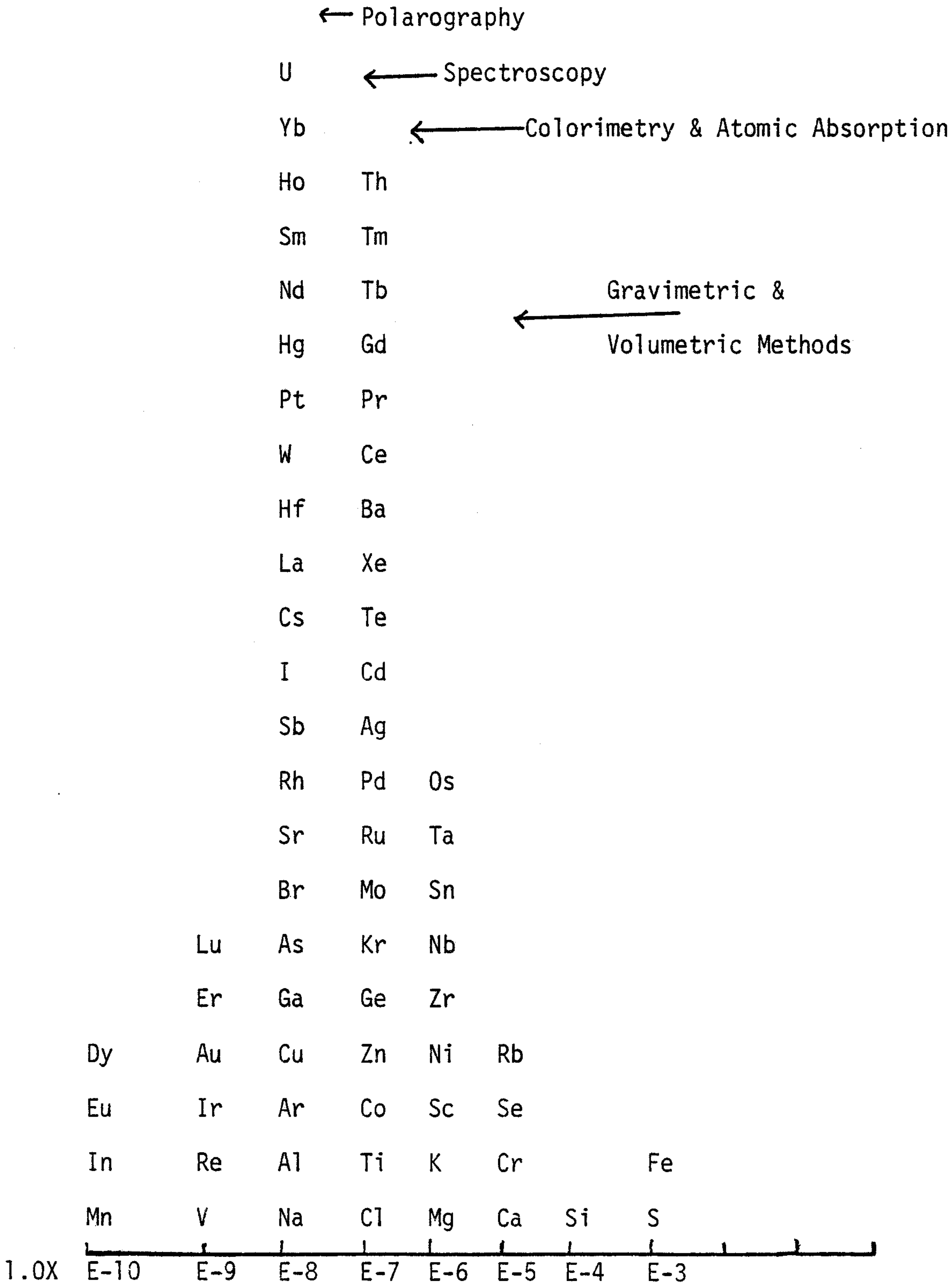
In quantitative analytical practices the general objective is the determination of some specific element or component. The method of analysis is chosen on the basis of the quality of the results desired. Some of the measures of the quality of the results are precision, accuracy, selectivity, sensitivity, detection limit and throughput of the method as well as the cost at which the results are obtained (44).

The objective of this study was to find a method of determining the optimum irradiation and decay times simultaneously so that instrumental thermal neutron activation analysis (INAA) can be carried out at conditions which give minimum matrix interference and hence optimum detection limit for any element of interest. These optimum conditions were to be available automatically once the required inputs of the optimization program have been specified.

1.2 NEED OF FINDING THE OPTIMUM CONDITIONS:

INAA is one of the most sensitive and nondestructive methods of single or multielement determination (see Figure 1). When Ge or Ge(Li) detectors with high resolution are used its selectivity is also very high. In analyses where activation of elements other than those to be determined occurs, the detection limit, accuracy and precision depend

Figure 1. Shows the detection limits of 79 elements irradiated for 1 hour at a thermal neutron flux of 10^{13} cm^{2} s^{-1} followed by gamma-ray spectroscopy using a 40 cm^{3} Ge(Li) detector (24).



			U					
			Yb					
			Ho	Th				
			Sm	Tm				
			Nd	Tb				
			Hg	Gd				
			Pt	Pr				
			W	Ce				
			Hf	Ba				
			La	Xe				
			Cs	Te				
			I	Cd				
			Sb	Ag				
			Rh	Pd	Os			
			Sr	Ru	Ta			
			Br	Mo	Sn			
		Lu	As	Kr	Nb			
		Er	Ga	Ge	Zr			
	Dy	Au	Cu	Zn	Ni	Rb		
	Eu	Ir	Ar	Co	Sc	Se		
	In	Re	Al	Ti	K	Cr		Fe
	Mn	V	Na	Cl	Mg	Ca	Si	S
1.0X	E-10	E-9	E-8	E-7	E-6	E-5	E-4	E-3

Detection limits for INAA(g/g)

on interfering gamma rays to a great extent (1-5, 12-16, 41).

Three types of possible interferences in instrumental activation analysis are: (i) interferences resulting from formation of the same radionuclide as that formed by an (n,γ) reaction of the element of interest. (Examples of this are fast neutron (n,p) and (n,α) reactions on an element one or two units higher in atomic number than the element of interest); (ii) interferences resulting from the formation of one or more radionuclides that emit gamma rays which overlap with the photopeak of interest and (iii) interferences resulting from Compton levels in the pulse height spectra caused by interactions of gamma rays of higher energy than the peak of interest upon which the peak of interest is superimposed. (See Figure 2a.)

Analyses carried out at optimum detection limit using INAA require that the effects of these interferences are reduced to a minimum. The first type of interference is brought under control by choosing irradiation positions where almost all fast neutrons are thermalized. The second problem is not often encountered when Ge or Ge(Li) detectors are used because of their high resolution, but when it does occur, the peaks can be resolved by mathematical methods implemented in the form of a subroutine of a spectral analysis program (30). The third problem is rather difficult to solve because it requires the proper choice of irradiation and decay times. This is the problem which calls for optimization of irradiation and decay time in INAA (1-18). An alternative approach to reduce Compton interference may be the use of chemical separation but in cases where a high throughput is required, separations consume time and may be too labourious.

There is also a general optimization usually carried out in neutron activation analysis to ensure that significant counts giving reasonable statistics for the peak of interest are obtained at reasonable count rates with minimum deadtime losses, pulse pile-up and spectral distortion. For these requirements, one optimizes the counting time, geometrical efficiency, the counting equipment and the data reduction method (22).

An example stressing the need for optimum decay and irradiation time is the analysis of atmospheric pollutants (27). In order to trace the pollution sources of aerosols and to determine how much the anthropogenic contribution is, it is necessary to determine the amount and elemental composition of atmospheric particulates and precipitation. For studies of sources, characteristics, atmoshperic transport processes and removal rates sensitive and multielemental analyses are required. Moreover, the method of analysis should be fast and easy because of the large masses of information required. Accurate analysis of airborne dust, dry deposition and rain water is difficult because of the complex nature of the samples and the low concentrations involved. Analytical techniques proposed for this purpose such as colorimetry, emission spectrometry, atomic absorption, flame photometry and polarography may or may not involve a dissolution step of the sample and subsequent chemical and instrumental steps. Since atmospheric particulate material is often to a great extent carbonaceous, it is largely insoluble in water. Ashing, in order to obtain complete dissolution, without loss of volatile elements is very difficult. Also the sensitivity and selectivity of most of these techniques is inadequate to cope with the complexity of environmental samples. Such analyses have been readily carried out by INAA using Ge (Li) or Ge gamma spectrometry and computer assisted data reduction (27, 28). The irradiation and decay time schedules which were used sometimes required several reirradiations and experience of handling the matrices involved (29). A method of easily finding these times for each element at optimum detection limit without having to do trial experiments is desirable.

1.3 THEORY OF INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS:

(I) Introduction:

The first part of INAA results in the production of radioactive nuclides formed when stable nuclides capture thermal neutrons. The completion of the analysis involves the detection and measurement of gamma rays emitted by desired nuclides among all those produced in the irradiated matrix. The radionuclides are characterised by both their decay schemes and their decay kinetics. The decay scheme summarizes the modes of disintegration and the energies of the radiations released. The kinetics describe the rates of disintegration of the radioactive nuclei.

The decay scheme is important in radioactivity measurements since it relates the number of radiations of a given kind and energy to the actual number of disintegrations of the particular nuclide. Energy values in gamma spectrometry provide qualitative information while the emission intensity of the gamma rays together with the decay kinetics provide both qualitative and quantitative information.

(II) Sample Activation:

If one considers a particular stable nuclide of an element that is capable of capturing a neutron, the number of radionuclides produced in a thin sample is given by the equation:

$$N = No \cdot \sigma \cdot \phi \cdot S/\lambda \qquad 1.$$

where No - is the initial number of target nuclei,

σ - is the reaction cross section (square cm),

ϕ - is the neutron flux (per square cm per second),

S - is the saturation factor and,

λ - is the decay constant (per second)

Sometimes the decay constant is expressed as:

$$\lambda = 0 \cdot 693/T \qquad 2.$$

where T - is the half-life of the radionuclide (in seconds).

The number of target nuclides for any particular isotope is given by the equation:

$$No = Na \cdot m \cdot J/Ma \qquad 3.$$

where Na - is the Avogadro number,

m - is the mass of element in irradiated sample (in grams),

J - is the isotopic abundance of the nuclide and

Ma - is the atomic weight of the element (in grams).

The saturation factor is given by the equation:

$$S = (1-e^{-\lambda Ti}) \qquad 4.$$

where Ti - is the irradiation time (in seconds).

The activity or rate of disintegration of the radionuclides just at the end of irradiation is given by the equation:

$$Ao = N \cdot \lambda \qquad 5.$$

(III) Detection of gamma rays:

Typically, the activated sample is counted at some decay time, Td, after the end of an irradiation period lasting for Ti. The activity after a decay time Td is:

$$A = Ao \cdot D \qquad 6.$$

where D - is the decay factor and is given by the equation:

$$D = e^{-\lambda Td} \qquad 7.$$

One seldom, in practice determines the actual disintegration rate of an induced activity, but rather one measures its counting rate under a given set of counting conditions. The counting rate depends upon the type of detector used, the size of the detector, the decay scheme of the radionuclide and the sample-to-detector geometry (solid angle subtended by the detector). If the counting period is Tc and the detector properties are characterised by the total detector efficiency (d(E)), the observed corrected total counts for a given gamma ray detected by Ge or Ge (Li) detector is obtained from the equation:

$$TCTS = I \cdot A \cdot d(E) \cdot (1 \cdot - e^{-\lambda Tc}) \cdot / \lambda \qquad 8.$$

where I - is the emission intensity of the gamma ray of interest with energy E.

If the counting time is much less than the halflife, then equation 8 can be recast in a different form:

$$TCTS1 = Tc \cdot A \cdot d(E) \cdot I \qquad 8a.$$

The counts arising from gamma rays as observed in the Multichannel Analyser (MCA) constitute a gamma spectrum. The spectral distribution of these counts is understood from considering the interaction of electromagnetic radiation with matter (see Table 1 and 2).

TABLE 1. Summary of the Interaction of Photons with Matter. (Ref. 22, Page 101)

Particle	Matter	Interaction		
		Elastic Collision	Inelastic Collision	Absorption
Gamma Photon	NUCLEUS	Negligible	Nuclear Resonance (e.g. Mossbauer Effect)	Photodisintegration
	ORBITAL ELECTRONS	Negligible	Compton Scattering	Photoelectric Effect
	FIELD	Negligible	Negligible	Pair Production for gamma energies > 1.02 Mev.

TABLE 2. Photon Escape and Origin of Peaks in Pulse-Height Spectra

Photon Escape Energy	Origin	Spectrum Peak Energy	Spectrum Peak Name
0	Total absorption	E	Photopeak
0.51Mev	Pair production and escape of one annihilation photon	E-0.51	First pair escape peak
1.02Mev	Pair production and escape of both annihilation photons	E-1.02	Second pair escape peak
$E''=E/(1+2\alpha)$	Compton 180° scattering	$E_{ce}=E/(1+\alpha/2)$	Compton edge
E" to E	Single Compton scattering	E_{ce} to 0	Single Compton distribution
0 to E	Multiple Compton scattering	E to 0	Multiple Compton distribution.

where $\alpha = E/mc^2$

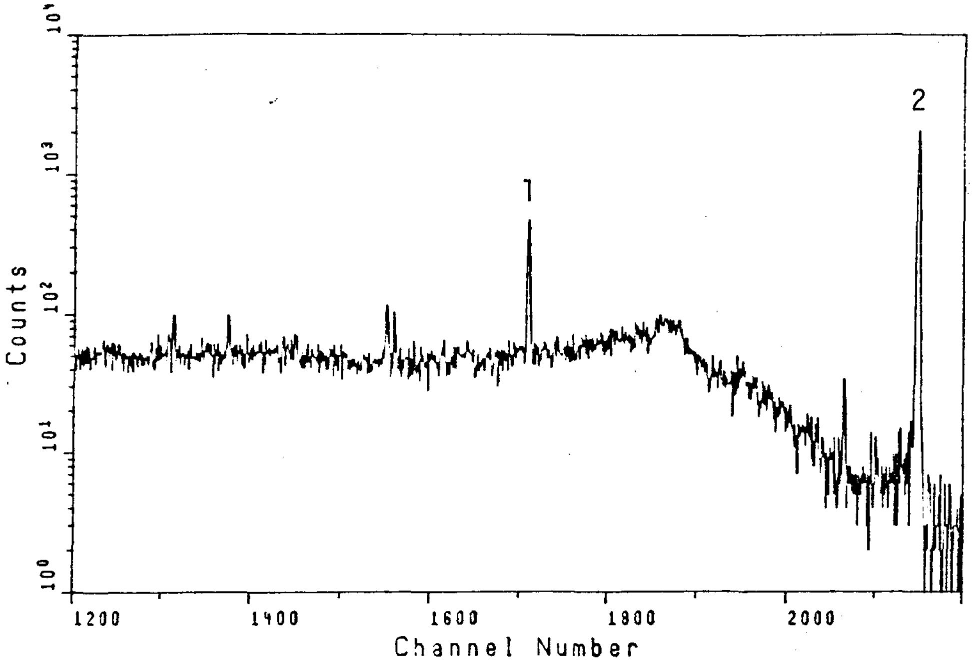
mc^2 = .511 MeV., i.e., electron mass energy and

180° = 180 degrees

A gamma ray entering a Ge or Ge(Li) detector can lose its energy through photoelectric interaction, or Compton scattering, or pair production (36-37). In photoelectric interaction, the incident photon loses all its energy to an electron of an atom which thus becomes ionized. The ejected electron can cause secondary ionization. The net charge induced in the detector is detected as a pulse whose magnitude is a function of energy or channel number in the MCA. The rate at which the pulses are produced depends on the disintegration rate of the sample. In Compton scattering, only part of the energy is imparted to the ejected electron. The scattered photon either undergoes further interaction or escapes from the detector. In pair production, photons with more than 1.02 Mev produce electron-positron pairs. This results in escape and .511 Mev peaks. The probability of photoelectric, Compton scattering and pair production are different and are functions of energy of the photon. Photoelectric interaction is predominant up to energies of about 1 Mev whereas Compton scattering is predominant up to energies of about 4 Mev. Pair production becomes the dominant form of interaction after 4 Mev (see Figure 2).

The detector characteristics (such as detector efficiency) depend on energy because all the interactions leading to registering the occurrence of an event are energy dependent. Usually, gamma ray spectrometry in INAA uses energies up to about 3 MeV so that the spectrum for a given gamma ray mainly consists of Compton events and total absorption events. This point of view (15) leads to the concept that for any sample, the spectrum is mainly made up of the sum of Compton events and photopeak events for all the gamma rays emitted by the sample.

The photopeak events approximately describe a gaussian peak and the Compton events approximately describe a rectangular function starting at the Compton edge. The Compton edge is the maximum energy a scattered photon can have. If CMP is the total Compton counts detected and PKC is the photopeak counts detected for a gamma ray of energy E whose emission rate is A, for a sample counted for Tc seconds, the total detector effici-



2a

Figure 2a. Part of a Spectrum of Y^{88} showing Compton interference.

1 = 1460 Kev peak of ^{40}K

2 = 1836 Kev peak of ^{88}Y

Peak 1 sits on the Compton background of Peak 2.

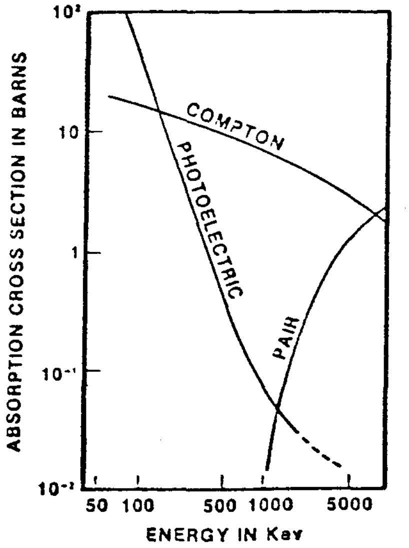


Figure 2b.
Variation of Photoelectric, Compton and Pair Production reaction cross section as a function of Energy.

ency is defined by:

$$d(E) = (PKC+CMP)/A \cdot Tc \qquad 9.$$

The photoefficiency (or photopeak efficiency), that is, the probability of detecting photopeak events during the counting interval is defined as:

$$Pf = PKC/(A \cdot Tc) \qquad 10.$$

If we substitute Pf for d(E) in equation 8, we obtain the photopeak count for a given gamma ray:

$$PKC = Tc \cdot No \cdot \sigma \cdot \phi \cdot S \cdot D \cdot I \cdot Pf \qquad 11.$$

The photofraction, that is, the fraction of events detected in the photopeak is:

$$f = PKC/(CMP + PKC) \qquad 12.$$

An alternative expression for the photopeak counts using equation 8 and 12 is:

$$PKC = TCTS \cdot f$$

or

$$PKC = Tc \cdot No \cdot \sigma \cdot \phi \cdot S \cdot D \cdot I \cdot d(E) \cdot f \qquad 12a.$$

and the Compton fraction is

$$cf = 1-f \qquad 13.$$

Using equation 8 and 13, the number of Compton counts for a given gamma ray then becomes:

$$CMP = TCTS \cdot cf \qquad 14.$$

or

$$CMP = Tc \cdot No \cdot \sigma \cdot \phi \cdot cf \cdot S \cdot D \cdot I \qquad 14a.$$

From equations 9, 10 and 12, an alternative expression for photopeak efficiency is:

$$Pf = d(E) \cdot f \qquad 15.$$

(IV) Quantitative determination in INAA

Three ways of determining the amount of an element in INAA use: (i) the absolute method or, (ii) comparator method or (iii) single comparator method.

The absolute method (28) is based on the determination of the weight of the element by means of the activation relationship 11. Rearranging equation 11 gives:

$$No = PKC/(Tc \cdot \sigma \cdot \phi \cdot S \cdot D \cdot I \cdot Pf) \qquad 16.$$

which is the number of radionuclides. Knowing the isotopic abundance and the atomic weight, this can be converted to the amount of element detected. However this method is rather crude because it assumes a steady flux of neutrons. The comparator method is much more accurate.

In the comparator method (28), the sample and a standard of the element of interest are irradiated under similar conditions. This avoids the need to directly use nuclear constants in finding the amount of the element. If A_u is the activity of the unknown sample and A_s is the activity of the standard weighing W_s grams, then the amount of the unknown sample is:

$$W_u = (A_u \cdot W_s)/A_s \qquad 17.$$

When multielement analysis involving shortlived nuclides or high throughput is required this method is inconvenient because it requires a standard for every element in the sample. A more convenient approach is to use a single comparator method (28).

In the Single Comparator method, the standard consists of a single element which if possible should have both longlived and shortlived activities (e.g. if Zinc is used we have the isotopes ^{69m}Zn (halflife 13.7 hr, γ-ray at 438.8 Kev) and ^{65}Zn halflife (243.7 d, γ-ray at 1115.5 Kev)).

An unknown concentration Cxy in a sample is calculated from the following equation:

$$C_{xy} = \frac{\psi_x \cdot (e^{-\lambda_x \cdot t_y}) \cdot S_s \cdot A_{xy} \cdot \varepsilon_s \cdot C_{1s}}{I_x \cdot (e^{-\lambda_s \cdot t_s}) \cdot S_x \cdot A_x \cdot \varepsilon_x \cdot C_{1y}} \qquad 18.$$

in which

$$\psi_x = \frac{\theta_s \cdot \sigma_s \cdot \gamma_s \cdot M_x}{\theta_x \cdot \sigma_x \cdot \Sigma\gamma_s \cdot M_s} \qquad 19.$$

$$I_x = \frac{\gamma_x}{\Sigma\gamma_x} \qquad 20.$$

where

x,y,s = subscripts for element, sample and standard, respectively

A = photopeak count rate

C_1 = weight

M = Atomic weight

λ = decay constant

S = Saturation factor

t = decay time

P = abundance of stable isotope

σ = effective cross-section for the irradiation position used

γ = abundance of selected gamma ray.

The ψ_X values depend only on the neutron spectrum in the irradiation position used. These can be determined experimentally for the isotope to be used. For isotopes with more than one prominent γ-ray, an average ψ_X value has to be obtained.

(V) Calculation of errors in INAA and detection limit

The error in the peak is usually obtained by assuming Poisson distribution of the counting rate (31). This assumption requires that the number of decaying nuclei of interest in the sample is very large and the probability of decaying in a given period is very small. The counting interval is chosen such that the total number of counts obtained (TCTS) is large. Under these conditions, if a radionuclide of interest emitting a characteristic gamma ray which is superimposed on Compton and room background, the standard deviation (31) of the peak counts for one run is:

$$\mathrm{SDEV} = \sqrt{\mathrm{CMP} + \mathrm{PKC} + \mathrm{BK}} \qquad 21.$$

where BK - is the room background,

CMP - is the Compton background under the peak and

PKC - is the peak area.

The % relative error (41) is given by:

$$S\% = SDEV.100/PKC \qquad 22.$$

The dependence of the detection limit (DL) on the height of the Compton continuum can be clearly seen from the formula introduced by Currie (37):

$$DL = (4.65 \cdot \sqrt{(BA)} + 2 \cdot 71)/PA \qquad 23.$$

where

BA - is the area on which the peak is situated (CMP + BK),

PA - is the peak area (PKC).

The base area is basically a function of the elemental composition of the matrix and its most important component is the Compton continuum.

1.4 GENERAL APPROACH TO OPTIMIZING IRRADIATION AND DECAY TIME

The general approach of optimizing irradiation and decay time is to define an analytical function which varies with both irradiation and decay time in such a way that the function is optimum when this type of interference is lowest for some set of irradiation and decay time. A nonzero optimum decay time only exists if the half-life of the interfering nuclide is less than half the halflife of the nuclide of interest

(7). When this requirement is not met, the optimum decay time is zero since waiting allows interfering activity to increase relative to the activity of interest. The procedure is to optimize the function and obtain the optimum irradiation and decay times as a set of times at which the function is optimum.

From a mathematical point of view, the problem becomes one of searching for extrema in a multidimensional space (45). This can be carried out by numerical or differential methods. It is preferable that the method chosen is easily programmed on a small computer.

Generally, differential methods find the stationary points of a continuous function at which the optimum is expected to be by evaluating its partial derivatives. For an unimodal function, the required stationary point is a maximum or a minimum. When the convexity of the function is known, the first partial derivatives adequately identify the extremum. The first derivatives are set equal to zero and solutions of the resulting equations when solved for the pertinent variables give the coordinates of the extremum. Methods for multimodal functions evaluate derivatives higher than the second so that stationary points such as points of inflextion and saddle points which are not extrema are distinguished. When it is not convenient to use differential methods the alternative is to use numerical methods. (In INAA all optimizations which have so far proved practical are numerical (5)). One starts with an initial approximation of the function and proceeds by generating a sequence of its approximations so that each of them is successively better than the previous one and nearer to the solution. This group of methods can be subdivided into arithmetic methods and logical methods. Arithmetic methods use an initial set of measurements to calculate the coordinates of the second set of measurements. This is continued until an optimum set is approached sufficiently well. The optimization usually proceeds by optimizing one variable at a time. An example of an arithmetic method is the Newton-Raphson method (2, 45). Logical methods select the positions of the second set of measurements by logical decisions. The second set of coordinates is accepted on the basis of the results of a logical comparison with the previous set of measurements. An example of a logical method is the simplex method (53).

1.5 REVIEW OF PREVIOUS WORK INVOLVING OPTIMIZING IRRADIATION AND DECAY TIME IN INAA

(i) Introduction:

The analytical functions used to find the conditions at which Compton interference is minimum should naturally be functions of Compton background. Some examples of such functions are detection limit (10, 12-16, 18), relative error (equation 22), and standard deviation (equation 21). In the optimization algorithm, the value accounting for Compton counts under the peak of interest can sometimes give problems if it is obtained from library spectra of interfering nuclides (8). Only sample matrices whose spectra are available can be analysed and samples must be measured under conditions which are identical to those under which the standard library spectra were measured. It is therefore preferable to use algorithms which do not need library spectra. A few examples of approaches used in previous attempts follow.

(ii) Examples:

1. Okada (1) theoretically considered optimizing the irradiation and decay time to improve the error and detection limit of the results of neutron activation analysis. His approach did not include information on Compton interference evaluation. He used an objective function proportional to the peak counts of interest, namely:

$$Y = S \cdot D \qquad 24.$$

where S - is the saturation factor and
D - is the decay factor.

The optimal conditions were obtained by using differential methods. This approach is rather limited because it ignores including Compton background - a feature which prompted the optimization. Furthermore, searching for optimum conditions using differential methods is rather difficult when there are several interferences as was pointed out by Isenhour and Morrison (2).

2. Isenhour and Morrison used an objective function defined as a ratio of the peak to the sum of the Compton background in the region of the peak and the peak counts, that is,

$$R = PKC/(PKC + \Sigma\ CMP) \qquad 25.$$

The fraction of the Compton events in the region of interest was obtained from standard spectra of each interfering nuclide. The Newton-Raphson method was used to find the optimum ratio with respect to one variable at a time. The starting point for the optimization was obtained by calculating a matrix of ratio values over the entire range of irradiation and decay time. The highest ratio was taken as the starting point.

3. Quittner and Montvai (3) used relative error as the objective function. They used differential methods to find the optimum conditions. They show results of sample calculations using the procedure for one interfering nuclide. It is likely that the method would have similar problems as those encountered by Isenhour and Morrison if tried for a more complex matrix.

4. Watterson (5) used the variance as the objective function. The optimization gave graphical solutions obtained from a graph of % relative standard deviation as a function of irradiation time only and another graph of % relative standard deviation as function of decay time only. Library spectra were used to find the fraction of Compton events in the region of the peak of interest.

5. In the method adopted by Guinn, a spectrum of the sample to be analysed is simulated for any given set of irradiation, decay and counting time on the basis of the fact that it is mainly composed of Compton events and photopeak events. Other peaks such as the escape or 511 peaks which

occur in the spectrum are also included. Then the % relative error and the detection limit for the peak of interest are calculated at these conditions. Several sets of conditions are tried and the one which gives the optimum % relative error to a level acceptable to the analyst is accepted as the optimum set of conditions. This method has been tested and used to determine the detection limits of several elements in INAA.

1.6 SIMPLEX OPTIMIZATION OF IRRADIATION AND DECAY TIMES

Only the algorithm adopted by Guinn meets both requirements of not needing library spectra and using appropriate functions. The others use rather restricted functions in terms of the number of interfering nuclides which can be considered (Ref. 1,3) or they need library spectra (Ref. 2,5). However the Guinn algorithm (called Advance Prediction) does not give optimum results automatically. The search for the optimum irradiation, decay and counting times requires selecting trial sets whose results the analyst has to examine. If the results are not satisfactory more trial sets have to be entered.

In this work, the advance prediction algorithm is used to generate the spectrum expected for the sample to be analysed, a figure of merit is defined,(that is, an analytical function of the property which must be optimized) and the modified simplex search method is used to simultaneously and automatically find the optimum irradiation and decay time.

CHAPTER 2

ADVANCE PREDICTION AND SIMPLEX ALGORITHMS:

2.1 ADVANCE PREDICTION:

The advance prediction algorithm uses a simple model of gamma ray pulse height spectra (41). This allows the calculation of the cumulative Compton continuum levels upon which the various photopeaks are superimposed. The method assumes that either the major constituents (major from the standpoint of levels of gamma ray emitting radionuclides produced) of the matrix are known, or that they have been established by means of a simple and rapid exploratory activation and gamma ray spectrometry measurement. In the original form of the Advance Prediction algorithm one starts by entering the sample composition followed by the entry of physical constants (e.g. atomic weight, Avogadro's number and isotopic abundances - to define the number of target nuclides) and the nuclear data constants (e.g. halflife, reaction cross section, emitted energies and their intensities). Then the detector characteristics are specified (in the form of equations of detector property as a function of energy). Finally the conditions (neutron flux, source-detector geometry, and decay, irradiation and counting times) of analysis are entered.

Equation 11a is used to calculate the photopeak counts and equation 14a is used to calculate the total Compton counts of each gamma ray. The Compton counts divided by the Compton edge gives the Compton distribution (Compton counts per unit of energy or Compton counts per channel) of a gamma ray. The cumulative Compton counts per channel is taken to be the sum of each gamma ray's Compton contribution in that channel.

In the original advance prediction, the main objective was to find the detection limit for any given set of conditions though one could obtain optimum times as well as optimum sample size. Hence, once the spectrum had been constructed, the detection limit or any other figure of merit (also called objective function or analytical function) to be evaluated is calculated. When optimization is the objective, several sets of times are entered and the one yielding the best results is chosen.

In this study, to search for optimum times, three sets of irradiation and decay time are entered after all the necessary parameters outlined above have been entered. A figure of merit is evaluated for each set. These sets of times together with their figures of merit define the initial simplex in three dimensions. The simplex algorithm is then used to find the optimum set of the figure of merit, irradiation time and decay time. Figure 3 gives a general outline of the Advance Prediction subroutines used in this study.

The detector properties required in the Advance Prediction algorithm depend on the quality of the simulated spectrum required. For a complete spectrum, one calibrates the detector for; (i) total detector efficiency, (ii) photofraction, (iii) Full Width at Half Maximum (FWHM), (iv) escape and .511 Mev peak efficiencies, and (v) energy (especially if a plot of the spectrum is desired).

Total detector efficiency and photofraction are necessary for photopeak and Compton count calculations as shown in equations 11 and 14

Figure 3. General outline of the subroutines necessary to predict a sample's spectrum in this study.

SAMDAT, PARAM and ENERGY are subroutines to enter sample composition, nuclear and physical constants (see Appendix B)

1 - Store emitting nuclide identifying code, halflife of the gamma emitting nuclidic state, the emitted energies and its saturation activity.

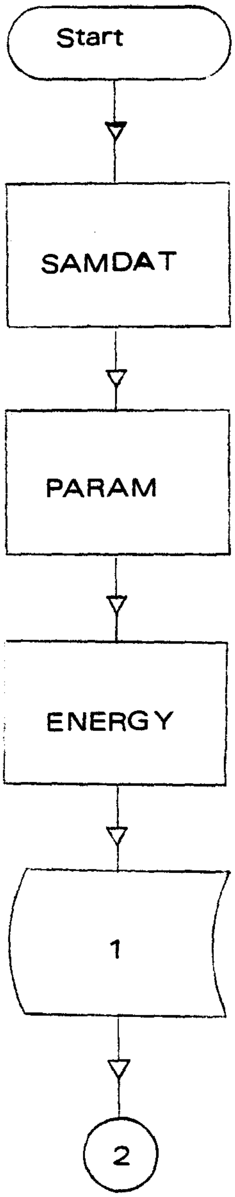
2 - Exit and enter the program or subroutine which needs a predicted spectrum (e.g. SIMPLEX·COM or GRID·COM).

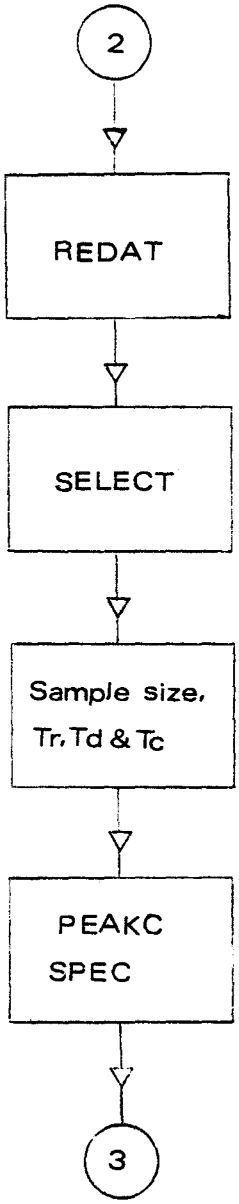
REDAT - Subroutine to read data stored at 1.

SELECT - Subroutine to select element and gamma energy of interest

PEAKC and SPEC - Subroutines to calculate the spectrum.

3 - Return to program needing the spectrum.


Start
SAMDAT
PARAM
ENERGY
1
2


2
REDAT
SELECT
Sample size,
Tr, Td & Tc
PEAKC
SPEC
3

respectively. The FWHM describes the number of channels a peak spans. This number is necessary when calculating the background under the peak. The escape (single and double) and .511 Mev peak efficiencies facilitate construction of realistic spectra when high energy gamma rays and positron annihilation are present. Energy calibration allows relating gamma ray energies to the peak positions (i.e. channel number) in the spectrum.

2.2 SIMPLEX ALGORITHM:

The simplex search method allows many factors to be varied at the same time to arrive at and track the optimum using rather simple decisions and coordinate calculations. This makes the method attractive for automated optimization. The method was first proposed by Spendley et al. (47). Its first application to analytical chemistry was by Ernst (49) who used it for instrumental optimization. Since then, there has been increased use of the method with various modifications aimed at improved convergence rates and precision. The Modified Simplex Method (MSM), an improved version of the original simplex algorithm (called the Basic Simplex Method) was used in this study. MSM was introduced by Nelder and Mead (48). In the original algorithm (BSM), the step size is fixed. When the step size is too small, it takes many moves to find the optimum whereas when it is too large, the optimum is determined with insufficient precision. In MSM, translation of the simplex is achieved by reflection, expansion, contraction and massive contraction. The step size is variable throughout the whole procedure. In this study, rules for the simplex moves are those to attain the minimum of the detection limit of the element of interest. The rules of performing these simplex moves can be understood by referring to Figures 4a and 4b.

RULES OF MODIFIED SIMPLEX METHOD.

A simplex is a geometrical figure defined by a number of points (n+1) equal to one more than the number of dimensions of the factor space(n). For the simplest multifactor problem, namely an optimization of two parameters, the simplex is a triangle. In the initial simplex BNW shown in Figure 4a, vertex B has the best response (that is, the lowest value of the objective function), W has the worst response and N has the next-to-worst response. P is the centroid of the line segment BN. It is logical to conclude that the response will probably be lower in the direction opposite to the point with the worst response (W). Therefore, the triangle is reflected through the point P to the point R, the reflected point. The coordinates of R are obtained according to the equation:

$$R = P + (P-W) \qquad 26.$$

The response at R can be lower than that at B, higher than that at B but lower than at N or higher than at N. Depending on which of the three possibilities is found to be true, the following steps are undertaken;

(i) If the response at R is lower than the response at B the simplex seems to move in a favourable direction. An expansion is therefore attempted by generating vertex E (the expanded point) by the equation:

$$E = P + \gamma\cdot(P-W) \qquad 27.$$

Figure 4a. Possible moves in the Modified Simplex Algorithm (Ref. 53)

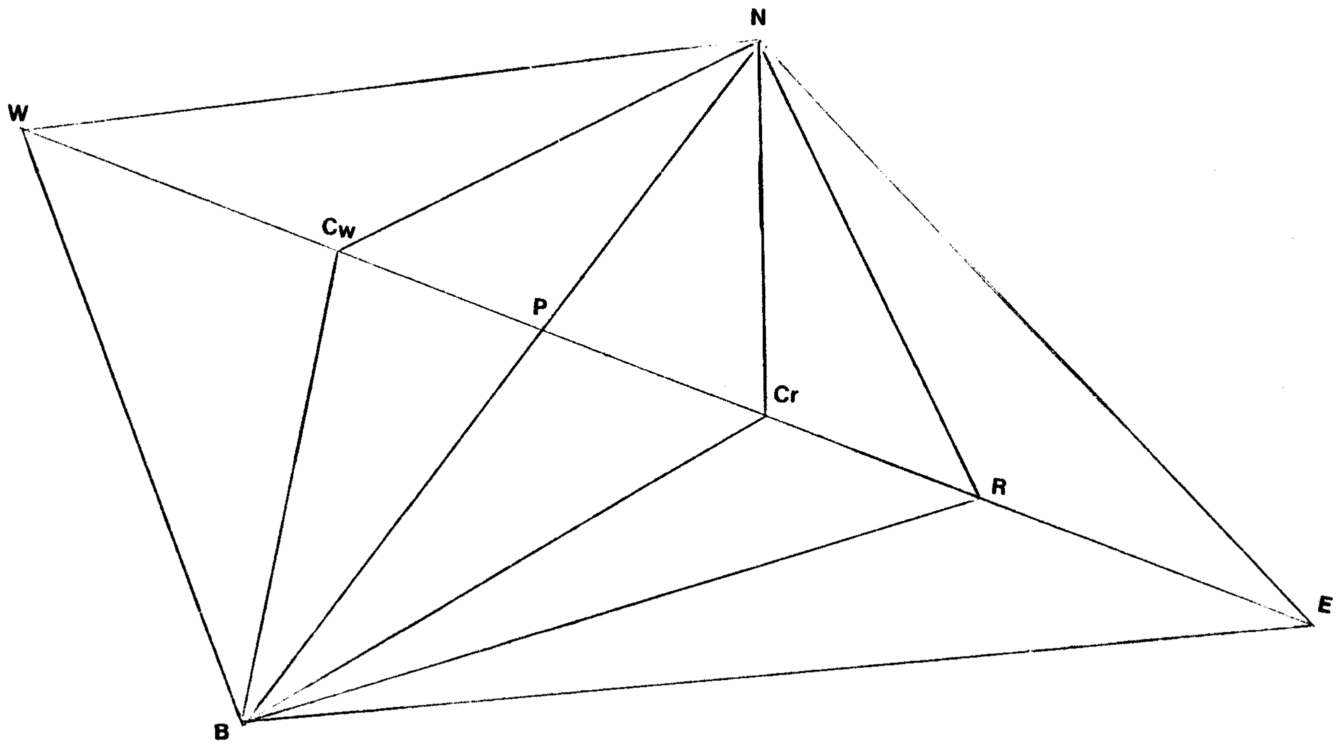
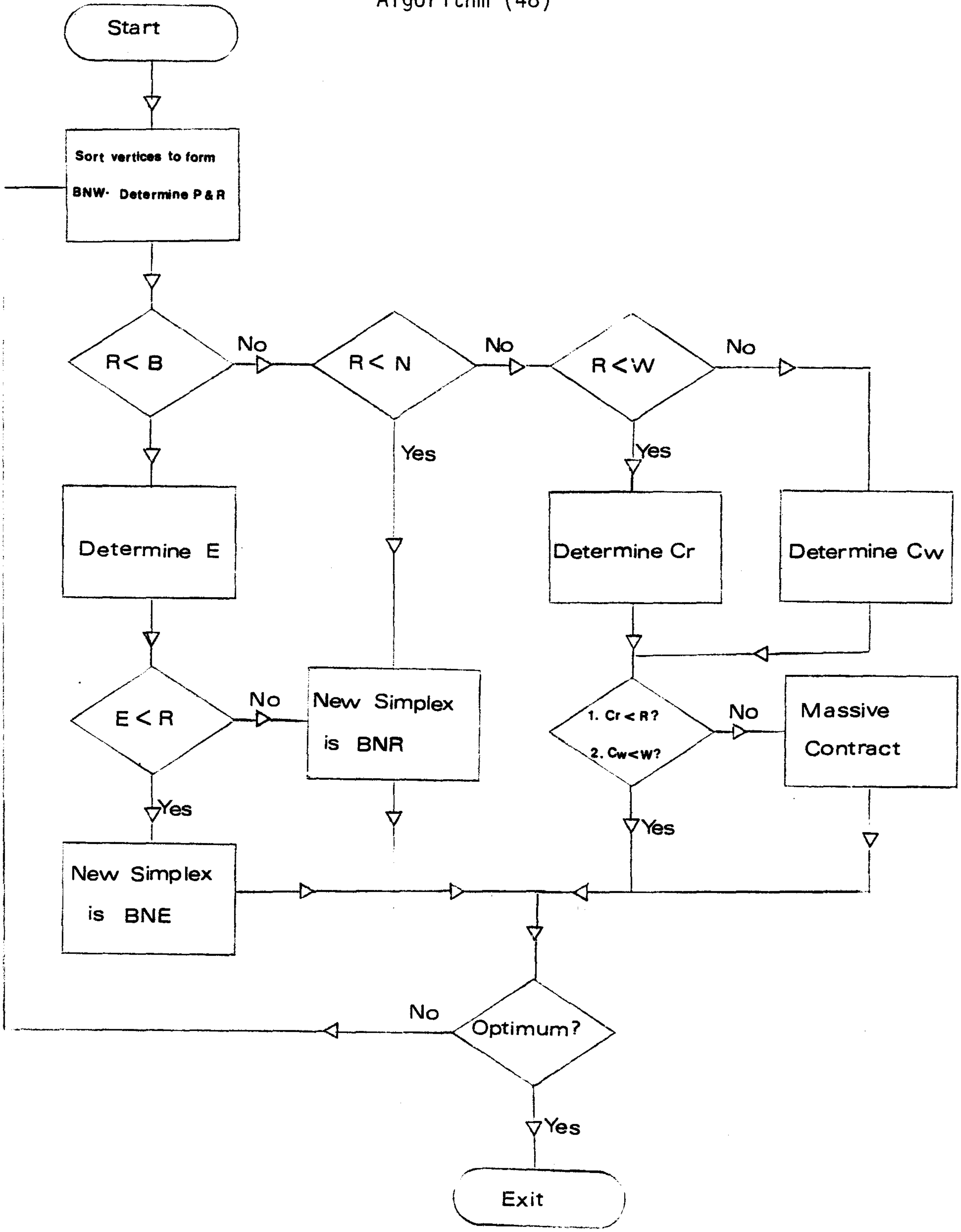


Figure 4b. Flowchart of the Modified Simplex Algorithm (48)



where γ is an expansion coefficient whose value is usually 2. If the response at E is also better than that at B, then E is retained and the new simplex becomes BNE. If not, the expansion has failed and the new simplex is BNR. One then proceeds to checking whether the optimum conditions have been attained. If the optimum has not been attained, one reflects the worst point and tries to reject it according to the prevailing simplex rules.

(ii) If the response at R is lower than the response at N but is greater than the response at B, the new simplex is BNR. No expansion or contraction is envisaged for the present simplex. One uses this new simplex to try to reject the worst point using rule (i) if the optimum has not been attained.

(iii) If the response at R is greater than that at N, the simplex has moved too far and should be contracted. The contraction can be around either the reflected point or the worst point. The decision of which point to contract around is based on the results of the comparison of the reflected point to the worst point.

1. If the response at R is lower than that at W, the new vertex Cr is best situated nearer to R. the contracted point is given by the equation:

$$Cr = P + \beta \cdot (P-W) \qquad 28.$$

where β is the contraction coefficient which is usually .5.

2. If the response at R is greater than that at W, the new vertex Cw is nearer to W and is given by the equation:

$$Cw = P-\beta\cdot(P-W) \qquad 29.$$

The new simplex is BNCr or BNCw and one proceeds in the usual way of trying to reject the worst point if the optimum has not been reached.

(iv) The failed contraction results if the response at Cr is greater than that at R or the response at Cw is greater than the response at W. Nelder and Mead recommend a massive contraction in which the size of the simplex is diminshed even further. This simplex move is sometimes referred to as shrinking the simplex. This is done by replacing N in the original simplex with P and retaining the current worst point (Cw or Cr). If for convenience we denote the point P as M, then the new simplex is BMCr or BMCw.

(v) If a vertex lies outside the boundaries of one or more of the factors, a very undesirable response is assigned to that vertex. The simplex will then be forced back inside the boundaries.

The simplex is halted when the standard deviation of the response at the three vertices becomes less than some predetermined value. Assessing that one is converging at the global optimum is done by carrying out several trials using different initial simplexes.

In this study, the simplex rules are used to calculate the values of irradiation and decay time for the different simplex translations. For each set of these times, defining a prospective simplex vertex, the advance prediction algorithm is used to generate a spectrum. This spectrum is used to evaluate the figure of merit. The figure of merit is used in the logical comparisons which lead to accepting or rejecting the point as a valid vertex of the new simplex.

CHAPTER 3

DETECTOR CALIBRATION

3.1 EXPERIMENTS

Several standard sources (see Table 3 and 4) were counted using a 45 cm^3 commercial cylindrical intrinsic Ge detector. The counting system consisted of a preamplifier, pulse pileup rejector, ratemeter, a spectroscopy amplifier with a 4 microsecond time constant and a multichannel analyser (see Table 5 and Figure 5). The source detector distance was defined by a rigid plastic shelfing structure with slots for a tray to hold the sample at various distances from the detector casing (see Figure 6). The sources were placed at the centre of the tray so that they were directly above the axis of the detector casing. All the sources were counted at a nominal distance of 3.7 cm from the detector casing. This distance was used because it was the lowest shelf slot and had therefore the maximum counting rate.

Three spectra were collected for each source. Three room background spectra were also collected for each counting period after counting the standard. These spectra were stored on a magnetic tape. A data routing system assembled by Burgess (38) was used to transfer spectra from the channel analyser to either the tape recorder for spectra storage or an Osborne 1 microcomputer. The microcomputer was used for spectral analysis. Spectral analysis was carried out using the program NAASYS (NEUTRON ACTIVATION ANALYSIS SYSTEM - Ref. 39).

TABLE 3. Standard Isotopes Used to Find Detector Characteristics

ISOTOPE	ISOTOPE ACTIVITY WHEN BOUGHT (in Microcuries)	GAMMA ENERGY OF INTEREST (in Mev)
Co-57	11.67 ±4.4%	0.12207 0.13643
Cs-137	10.57±3.7%	0.66162
Na-22	10.16±3.7%	0.51100 1.27454
Mn-54	10.26±3.7%	0.83481
Co-60	10.74±1.9%	1.17323 1.133251
Ba-133	11.36±4.8%	0.35586
Y-88	11.46±5.0%	0.89802 1.83601

TABLE 4. Energies used for energy calibration in Ra-226

PEAK NUMBER	ENERGY IN MEV
1	0.2952
2	0.3519
3	0.6095
4	0.7684
5	0.9341
6	1.1201
7	1.2381
8	1.3777
9	1.7296
10	1.7645
11	1.8474

TABLE 5. COUNTING SYSTEM COMPONENTS

INSTRUMENT	MODEL	SUPPLIER
Passivated Hyperpure Germanium Detector	CS-A31C(1.8)	Aptec/NRD
Spectroscopy Amplifier	2010	CANBERRA
Linear Ratemeter	1481	CANBERRA
Livetime Corrector/ Pileup Rejector	1468	CANBERRA
Multichannel Analyser	8100	CANBERRA
Reference Pulse Generator	1407	CANBERRA
Data Cassette	8410	TECHTRAN

Figure 5. Gamma Spectrometry System

A : Tape, B : Microcomputer & C : Teletype

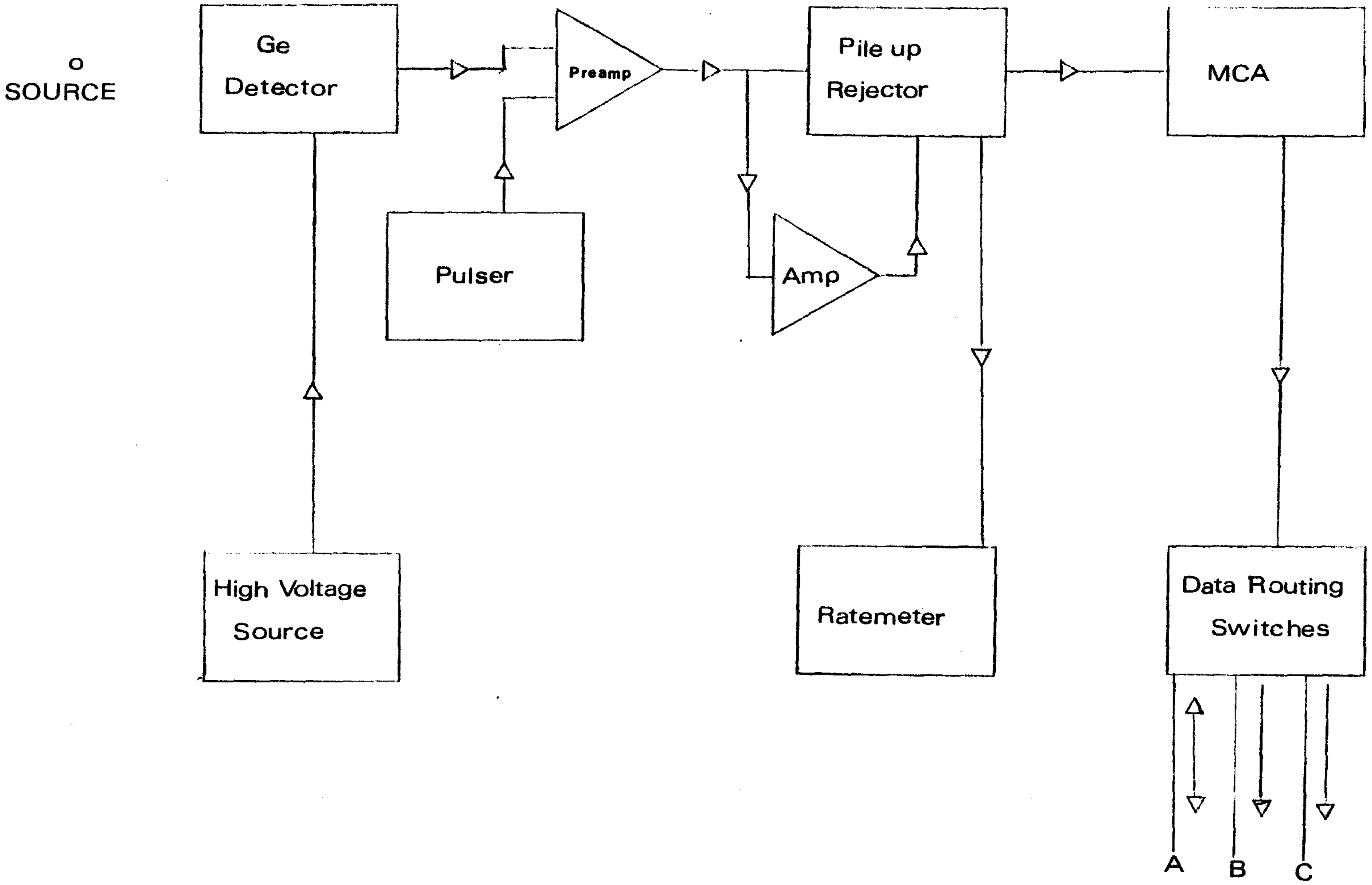
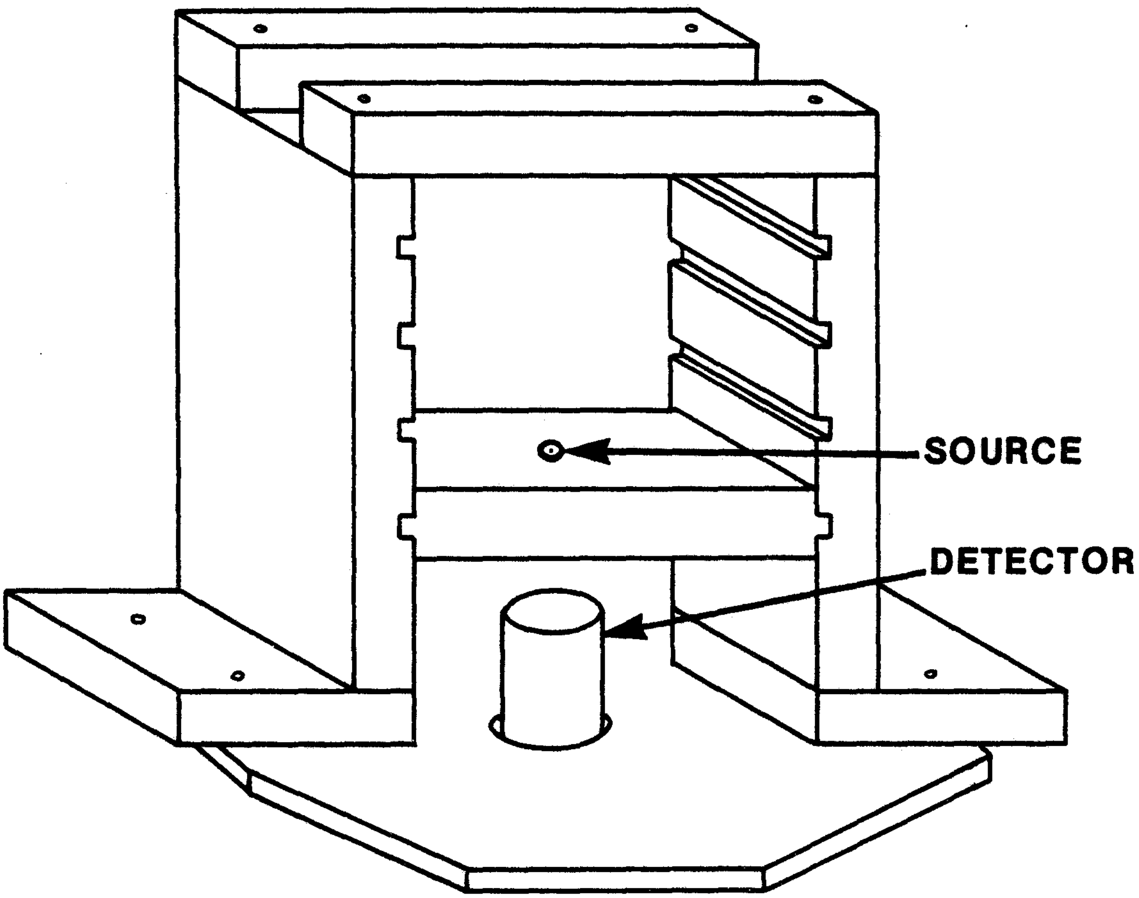


Figure 6.

Source-Detector Configuration



3.2 DATA REDUCTION:

(I) ENERGY CALIBRATION:

When a sample's spectrum is obtained, a spectral analysis program in INAA essentially calculates two parameters from the spectrum; viz.; the energies and the areas of the peaks. Peaks in the pulse height channel analyser are assigned to channels rather than energies. It is therefore necessary to find the relationship between channel numbers and peak energies before performing spectral analysis on the sample's spectrum. Establishing this relationship is called energy calibration. (It should be mentioned that energy calibration is not limited to the case of using a spectral analysis program. Rather, it is part of the procedure of obtaining qualitative information in INAA.)

The relationship between energy and channel number is defined by a polynomial function (27) of the form:

$$E = a_0 + a_1 \cdot C + a_2 \cdot C^2 + a_3 \cdot C^3 + \ldots \qquad 30.$$

where E - is the peak energy,

C - is the channel number at the peak centroid and

a_i - stands for calibration constants.

The calibration constants are the values which are sought for during energy calibration. This is performed by using standard sources with known energies. The channel numbers are obtained from the spectra of the standards. The two parameters (energy and channel number) are then used in a least squares program to find the required constants.

The polynomial nature of the peak energy - channel number relationship is due to the non-linearity of the spectroscopy amplifier. If the amplifier response can be defined by a linear function, only two gamma energies are needed for calibration. Otherwise more energies are needed. The sources can be:

(i) several single energy emitting nuclides in the form standards of single nuclides, or

(ii) several single or multiple energy nuclides in the form of a mixture of nuclides (e.g. Ra-226), or

(iii) a single nuclide emitting at least two gamma ray energies (e.g. Co-60).

From the experimental point of view it is more convenient to use a source for which only one spectrum is needed to obtain several peaks. This favours type (ii) and (iii) sources.

The NAASYS program has an energy calibration subroutine which makes available the constants to the spectral analysis subroutine. A standard of Ra-226 was used for energy calibration.

(II) CORRECTIONS REQUIRED ON INAA SPECTRA

1. Pulse pile-up: This happens when a pulse occurs in the detection system sufficiently soon after a preceding one to combine their heights. This is prevalent at high counting rates. Instead of two pulses, the system registers one pulse with an energy somewhere between the individual components and their sum. The effect is to remove counts from the peaks and change the shape of the spectrum. The pulse pile-up rejector was used to reject such peaks automatically in order to reduce spectral distortion.

2. System deadtime losses: System deadtime refers to the fact that a multichannel analyser requires a period of time to process a signal due to a detected event. During this time, other signals are lost. The fraction of counts lost in all the peaks of significant intensity are the same. To correct for system dead time, the reference pulse generator was used to generate pulses at a predetermined frequency of 60 Hz and these pulses (of a chosen amplitude and therefore chosen channel) were sent to the preamplifier. A certain fraction of these pulses were lost through the dead time phenomenon so that the pulser peak counts observed were less than those determined by the product of the pulser frequency and counting interval. The dead-time correction factor was obtained by using the equation:

$$r = Pex/Pob \qquad 31.$$

where Pex - is the expected pulser counts and

Pob - is the observed pulser counts.

$$Pex = 60 \cdot Tc \qquad 32.$$

3. Room Background: The determination of TCTS (equation 8) for a given gamma ray requires the integration of all channel counts up to the high energy peak boundary channel (obtained from NAASYS). Since there is always contribution from room background for a spectrum, this was subtracted from the obtained integral.

(III) PHOTOPEAK EFFICIENCY (Pf(E)):

After energy calibration, NAASYS was used to calculate the peak areas of spectra of the standard sources. The peak areas were corrected for dead-time losses and used to calculate the photopeak efficiencies for each gamma energy according to equation 10. The photopeak efficiency as shown in equation 16 is an important quantity for quantitative analysis. In this study it was used to check the validity of the relationships of photofraction versus energy and total detector efficiency versus energy by using equation 10. It was necessary to check these relationships because of the inherent approximations used to obtain them (see below).

(IV) DETECTOR EFFICIENCY (d(E)):

The total detector efficiency was obtained from standard sources with one or two gamma rays by using equation 9. For sources with two gamma rays, either the efficiency was assigned to the average energy (Co-57 and CO-60) or the gross count (TCTS) was decomposed to obtain a value at one energy (Y-88 and Na-22) (see Appendix A).

(V) PHOTOFRACTION (f(E)):

The photofraction was obtained by using equation 12 if there was only one gamma ray emitted by the nuclide (e.g. Cs-137). For nuclides emitting two gamma rays close together, the average total detector efficiency and the average photofraction were used (see Appendix A). In the case where the two gamma rays were too far apart as in Y-88, a regression program (43) was used to find the photofraction flanked by photofractions which could directly be obtained by equation 12. For example the photofractions of Cs-137 (.662 Mev), Mn-54 (.835 Mev) and Co-60 (1.253 Mev) were used in the regression program to find the photofraction of the .898 Mev gamma ray of Y-88. Once this was found, since its peak counts could be obtained from the spectrum, use of equation 12 gave the total counts due to the 0.898 Mev gamma ray. The photofraction of the other gamma ray (1.836 Mev) was found by first determining the total counts (as in Appendix A, Case 2) followed by application of equation 12 since the peak counts are known from the use of NAASYS.

(VI) FULL WIDTH AT HALF-MAXIMUM (FWHM):

The peak width in gamma spectrometry is expressed as FWHM (27, P178). Theoretically the following relationship exists between resolution and energy:

$$(\mathrm{FWHM})^2 = k \cdot f \cdot \varepsilon \cdot E + (\rho)^2 \qquad 32.$$

where k - is a proportionality constant;

f - is the Fano factor

ε - is the energy required to produce one electron-hole pair (2.98 eV for Ge);

E - is the energy of the gamma photon and

ρ - is the contribution to the resolution by electronic factors (noise from detector leakage current, preamplifier, etc.).

The FWHM will become practically constant below a certain energy, depending on the quality of the detector and associated electronics.

The method of Zimmerman (64) was used to determine FWHM at various gamma energies. In this method, the peak is considered as a gaussian peak. A linearised form of the peak is obtained by plotting the natural logarithm of the ratio $Y(x-1)/Y(x+1)$ against x; where x is the channel number and $Y(x)$ is the number of counts due to the peak (i.e. background corrected counts) in channel number x. The slope of the linear part of the graph is given by the equation:

$$\text{slope} = 2\cdot/(\sigma)^2 \qquad 33.$$

The FWHM is given by the equation:

$$\text{FWHM} = 2.355\cdot\sigma \qquad 34.$$

Linear regression was used to obtain the linear part each linearised gaussian curve (peak) for several standard sources.

3.3 RESULTS

Tables 6-9 show the photopeak efficiency, total detector efficiency, photofraction and FWHM respectively. Since these parameters are functions of energy, their proper characterisation required finding their functional relationships with energy. The relationships were sought for using a regression program (43). The function giving the best correlation for each characteristic was chosen. These functions together with their experimental values used to generate them were plotted (see Figures 7-10). Table 10 shows the function of each characteristic.

TABLE 6. Table of photoefficiency of several gamma energies.

Source	Energy (Mev.)	Photopeak Efficiency (%)
Co-57	0.122	1.064±0.048
Ba-133	0.356	0.396±0.019
Na-22	0.511	0.248±0.009
Cs-137	0.662	0.195±0.007
Mn-54	0.835	0.153±0.006
Y-88	0.898	0.139±0.007
Co-60	1.173	0.106±0.002
Na-22	1.275	0.096±0.003
Co-60	1.333	0.093±0.002
Y-88	1.836	0.068±0.003

These photoefficiencies were calculated by obtaining the mean peak count for three experimental runs of counting the Source. The results in this Table were used in a regression program to derive the first function in Table 10. This function was used together with the data in this Table to generate Figure 7.

Figure 7. Graph of photopeak efficiency as a function of energy. The solid curve was obtained by using the first equation in Table 16 and points were obtained by using the data in Table 6.

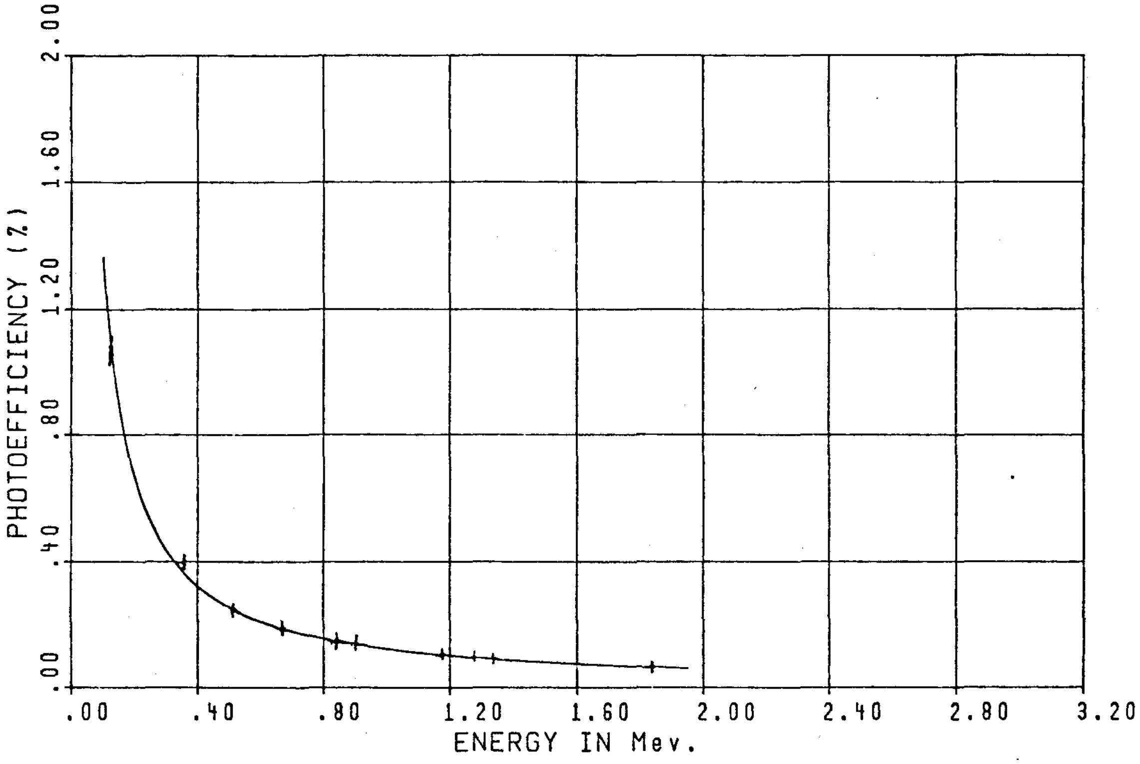


TABLE 7. Table of Total Detector efficiency for several gamma rays.

Source	Energy (Mev.)	Total Detector Efficiency (%)
Co-57	0.124 *	1.76±0.08
Na-22	0.511 **	1.17±0.04
Cs-137	0.662	1.14±0.04
Mn-54	0.835	1.06±0.04
Co-60	1.253 *	0.97±0.02
Y-88	1.836 **	0.95±0.05

N.B. The energies marked with one asterisk are average values and these efficiencies were obtained by the Case 1 method (see Appendix A). The energies marked with two asterisks were obtained by using the method outlined in case 2.

These total detector efficiencies were used in a regression program to generate the second function in Table 10. This function together with the data in this Table were used to obtain Figure 8. The total detector efficiencies shown in the Table are the mean of three experimental measurements.

Figure 8. The graph of Total Detector Efficiency as a function of energy. The solid curve was obtained by using the second equation in Table 16 whereas the points are from Table 7.

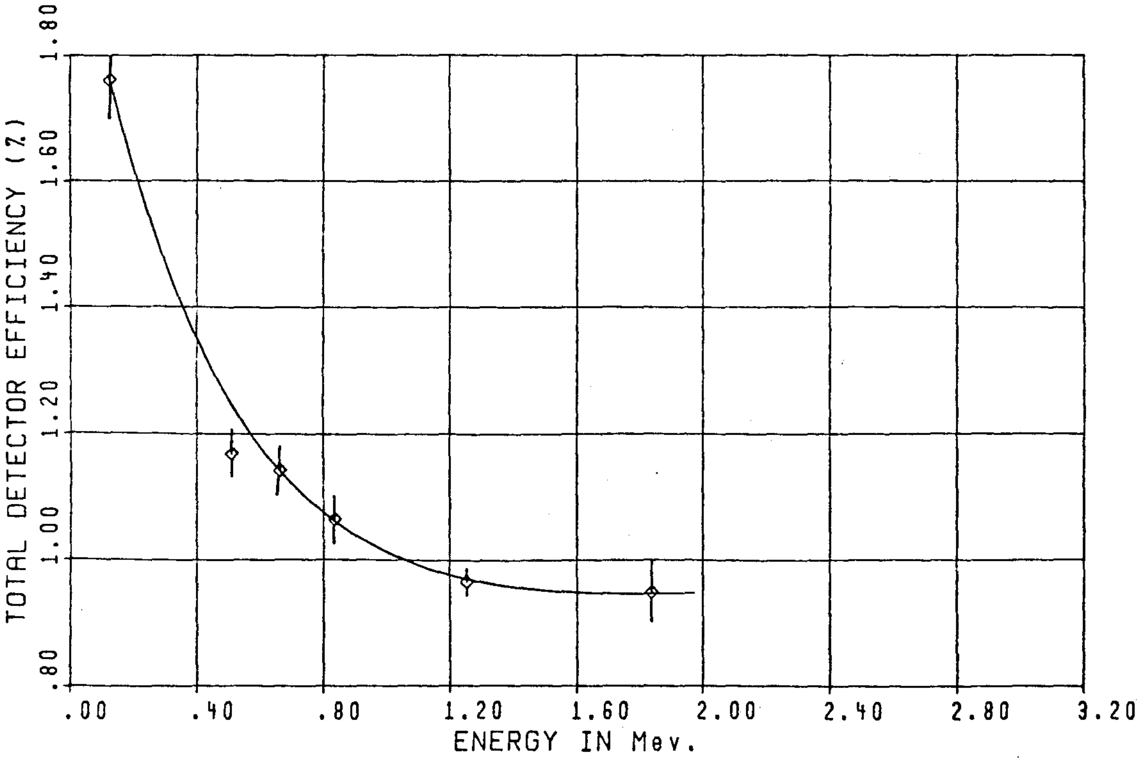


Figure 9. Photofraction as a function of Energy. The solid curve was obtained by using the third equation in Table 16 whereas the points represent results in Table 8.

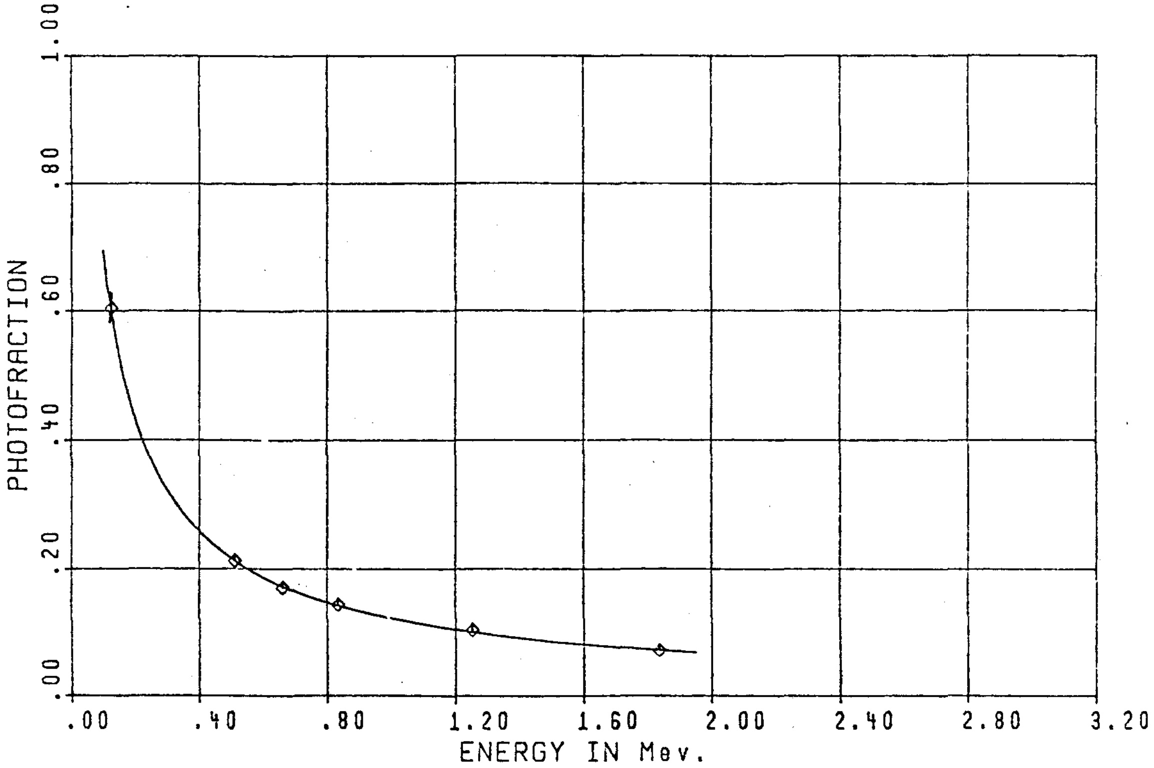


TABLE 8. Table of Photofraction of several gamma energies.

Source	Energy (Mev.)	Photofraction (unitless)
Co-57	0.124 *	0.603±0.027
Na-22	0.511 **	0.212±0.008
Cs-137	0.662	0.170±0.006
Mn-54	0.835	0.144±0.005
Co-60	1.253 *	0.103±0.002
Y-88	1.836 **	0.077±0.004

N.B. The photofractions of energies marked with an asterisk are the mean energy whereas those marked with one asterisk are obtained by first determining the photofraction of the other gamma energy's photofraction first.

These photofractions were used in the regression program to generate the third function in Table 10. This function was together with the data in this Table were used to generate Figure 9. The photofractions as seen in this Table are the mean of three experimental measurements.

Figure 10. The Square of FWHM as a function of Energy. The points represent the results in Table 9 whereas the solid curve was obtained by using the fourth equation in Table 16.

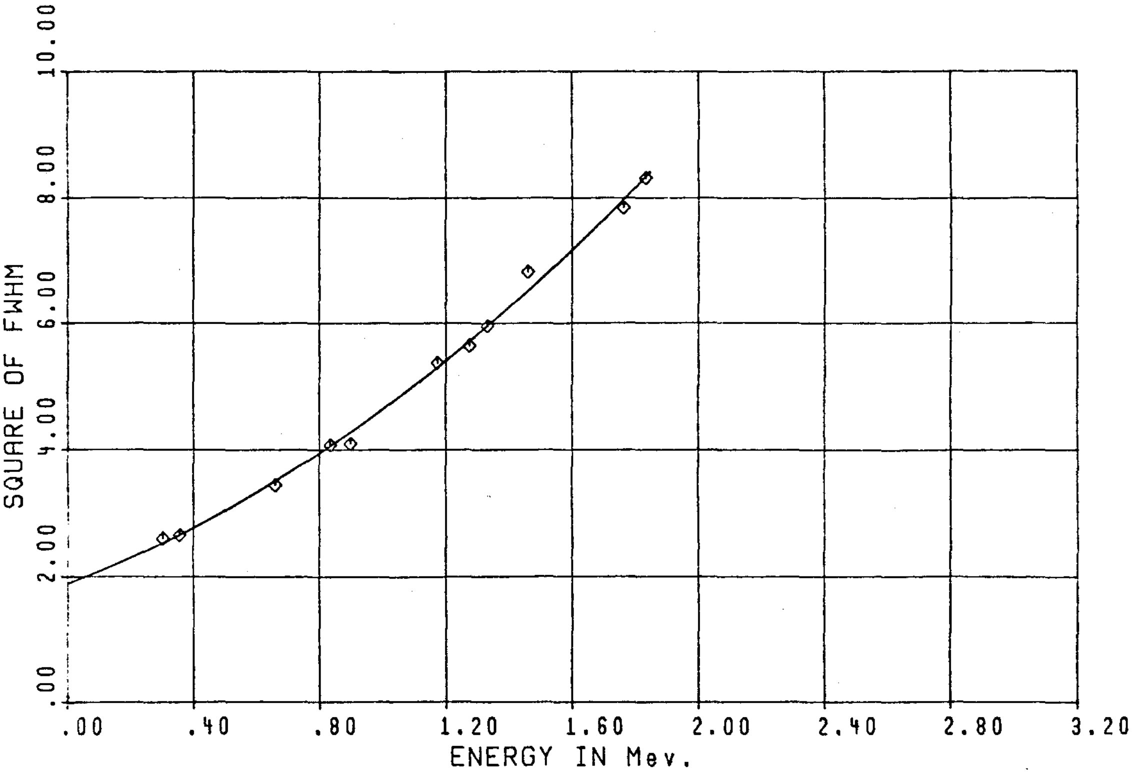


TABLE 9. Table of Full Width Half-Maximum for several gamma energies.

Source	Energy (Mev.)	FWHM (channels)	Square of FWHM
Ba-133	0.302	1.61±0.06	2.608±0.004
Ba-133	0.356	1.63±0.06	2.659±0.004
Cs-137	0.662	1.85±0.05	3.440±0.003
Mn-54	0.835	2.02±0.05	4.078±0.003
Y-88	0.898	2.02±0.05	4.079±0.003
Co-60	1.173	2.32±0.02	5.3796±0.0005
Na-22	1.275	2.40±0.04	5.747±0.002
Co-60	1.333	2.42±0.03	5.831±0.001
K-40	1.46	2.61±0.01	6.825±0.0001
Ra-226	1.764	2.80±0.01	7.8456±0.0002
Y-88	1.836	2.88±0.08	8.318±0.004

The results in thie Table are the mean of three analyses and were used to determine the fourth function in Table 4. The function together with these results, were used to obtain Figure 10.

Table 10. Table of functions used to define the detector properties.

Property	Function	Correlation
Photo efficiency -(%)	$Pf(E) = e^{-(2.084 + 1.046 \ln E)}$	0.9988±0.0007
Total Detector Efficiency (%)	$d(E) = 0.9895-0.513 e^{-E} + 1.57 (e^{-E})^2$	0.9999±0
Photo fraction -(unitless)	$f(E) = e-(2.1 + 0.854(\ln E + 0.043(\ln E)^2)$	0.9998±.0001
Square of Full Width at Half Maximum -(channel)^2	$(FWHM)^2 = 1.885 + 1.85E + 0.907E^2$	0.9981±0.0013

N.B. 1. To convert FWHM to energy we need an energy calibration factor. This is 0.844 so that FWHM is in Kev;

$$FWHM = 0.844 \sqrt{1.885 + 1.85E + 0.907}E^2$$

2. In the equations above E is the energy in Mev.

CHAPTER 4

4.1 SIMPLEX OPTIMIZATION BY ADVANCE PREDICTION PROGRAM

Once the detector parameters were determined, the simplex optimization by advance prediction program was written. An Osborne I microcomputer with a random access memory of 64K was used. The microcomputer used a Facit 4510 serial matrix printer and a 7470A Hewlett-Packard plotter. All routines were written in Microsoft Fortran.

The programs were developed as three different major modules, viz; (i) a program with routines for entering the sample composition, physical and nuclear constants and calculating the saturation activity for each gamma ray in a gram of sample; (ii) a modified simplex optimization program which accesses the saturation activity calculated in the above program. After accessing this data, the program enters the limiting times of the search region, the sample size and calls the spectrum predicting routines. Finally the detection limits are determined and used in the simplex moves. (iii) the final program uses the data from the first program just like the simplex program. After reading the data the spectrum predicting routines are used to find the detection limits for predetermined sets of irradiation and decay time. Each set of irradiation time, decay time and detection limit define a point in three dimensions. The set of all points define a three dimensional grid of detection limit.

The plot of the grid gives the topography of the response function (42). This shows whether the simplex program converges in the region of minimum detection limit. It also shows the kind of response surface that is being considered.

4.2 STRATEGY OF EVALUATING THE PROGRAM

To evaluate the program, an airborne particulate sample composition compiled from the literature (29) was used. The composition was made to consist of a representative set of halflives and compositions usually found in airborne particulate analysis; that is, the sample had isotopes of short, medium and long halflife as well as major and minor constituents (in terms of composition) in these halflife ranges (see Table 11). With this kind of composition, for each range of halflife, the simplex optimization program was implemented for the elements in Table 12. Three initial simplex were started in different regions for each element to observe its convergence region. Then a grid plot for each element was obtained.

4.3 RESULTS

Table 13 shows the boundary conditions for the elements optimized for and Table 14 shows the optimum times obtained. Table 15 shows the starting region of the simplex for three elements, their convergence region. Figures11-13 show examples of topography of the search space.

TABLE 11. Table of sample composition used to examine program (Ref. 29)

Element #	Wt. of Element in Sample (ng/m^3)	Element
1	1161.5	Na
2	2659.0	Mg
3	4752.3	Al
4	3479.5	Cl
5	1867.5	K
6	3227.5	Ca
7	1.55	Sc
8	517.5	Ti
9	13.5	V
10	40.9	Cr
11	201.0	Mn
12	6363.0	Fe
13	5.2	Co
14	292.0	Cu
15	410.5	Zn
16	31.4	As
17	6.86	Se
18	465.5	Br

Table 12 Table of Elements Optimized for Using the Simplex Program

Isotope	Gamma Energy (Kev)	Halflife (Minutes)
^{28}Al	1778.8	2.24
^{51}Ti	320.0	5.79
^{27}Mg	843.7	9.46
^{52}V	1434.2	3.75
^{38}Cl	1642.4	37.3
^{56}Mn	846.6	154.74 (2.579 hrs.)
^{69m}Zn	438.6	828.0 (13.8 hrs.)
^{76}As	559.1	1578.0 (26.3 hrs.)
^{82}Br	776.5	2116.8 (35.28 hrs.)
^{51}Cr	320.07	3989.0 (27.70 dys.)

Table 13 Boundary Conditions used for Optimization

Isotope	Tc in min.	TDMIN in min.	TDMAX in min.	TRMIN in min.	TRMAX in min.
^{28}Al	5.0	0.2	100.0	0.2	480.0
^{51}Ti	5.0	0.2	100.0	0.2	480.0
^{27}Mg	5.0	0.2	100.0	0.2	480.0
^{52}V	5.0	0.2	100.0	0.2	480.0
^{38}Cl	5.0	0.2	100.0	0.2	480.0
^{56}Mn	5.0	0.2	480.0	0.2	480.0
^{59m}Zn	100.0	0.2	7200.0	0.2	480.0
^{76}As	60.0	0.2	7200.0	0.2	480.0
^{82}Br	60.0	0.2	7200.0	0.2	480.0
^{51}Cr	60.0	0.2	72000.0	0.2	480.0

N.B. The simplex halted when the Standard deviation of the vertices was less than 1.0×10^{-6}.

Table 14. Table of Optimum Times for Several Elements

Isotope	$\tau_{\frac{1}{2}}$	Optimum TDm min.	Optimum Trm min.	Energy in Kev
^{28}Al	2.24 min.	0.2	5.5	1778.8
^{52}V	3.75 min.	0.20	22.91	1434.2
^{51}Ti	5.79 min.	0.20	21.07	320.0
^{27}Mg	9.46 min.	3.89	37.66	843.7
^{38}Cl	37.3 min.	0.20	54.7	1642.4
^{56}Mn	2.579 hrs.	20.38	300	846.6
^{69m}Zn	13.8 hrs.	645.0 (10.75 hrs.)	480	438.6
^{76}As	26.3 hrs.	1167.4 (19.28 hrs.)	480	559.1
^{82}Br	35.28 hrs.	1275.0 (21.25 hrs.)	480	776.5
^{51}Cr	27.7 dys.	25100.0 (18.125 dys.)	480	320.07

Table 15a. Table of Initial and Final Simplexes of ^{76}As

Run #	Vertex	TC in Min.	TR in Min.	Type of Simplex
1	1	1600.0	450.0	Initial
	2	40.0	400.0	
	3	6000.0	300.0	
2	1	3500.0	250.0	Initial
	2	7000.0	450.0	
	3	10.0	100.0	
3	1	300.0	300.0	Initial
	2	150.0	170.0	
	3	2.0	100.0	
1	1	1168	480.0	Final
	2	1172	480.0	
	3	1169	480.0	
2	1	1157	480.0	
	2	1157	480.0	
	3	1157	480.0	
3	1	1174	480.0	
	2	1178	480.0	
	3	1175	480.0	

N.B. (i) TD = Decay time (in Minutes)

(ii) TR = Irradiation time (in Minutes)

(iii) The optimum times are rounded off on output of results to give four digits (original were results in Exponential for mat).

Figure 11. Grid Plot of the Response Surface of ^{76}As (Tc = 60.0 Minutes)

* Point of Simplex convergence.

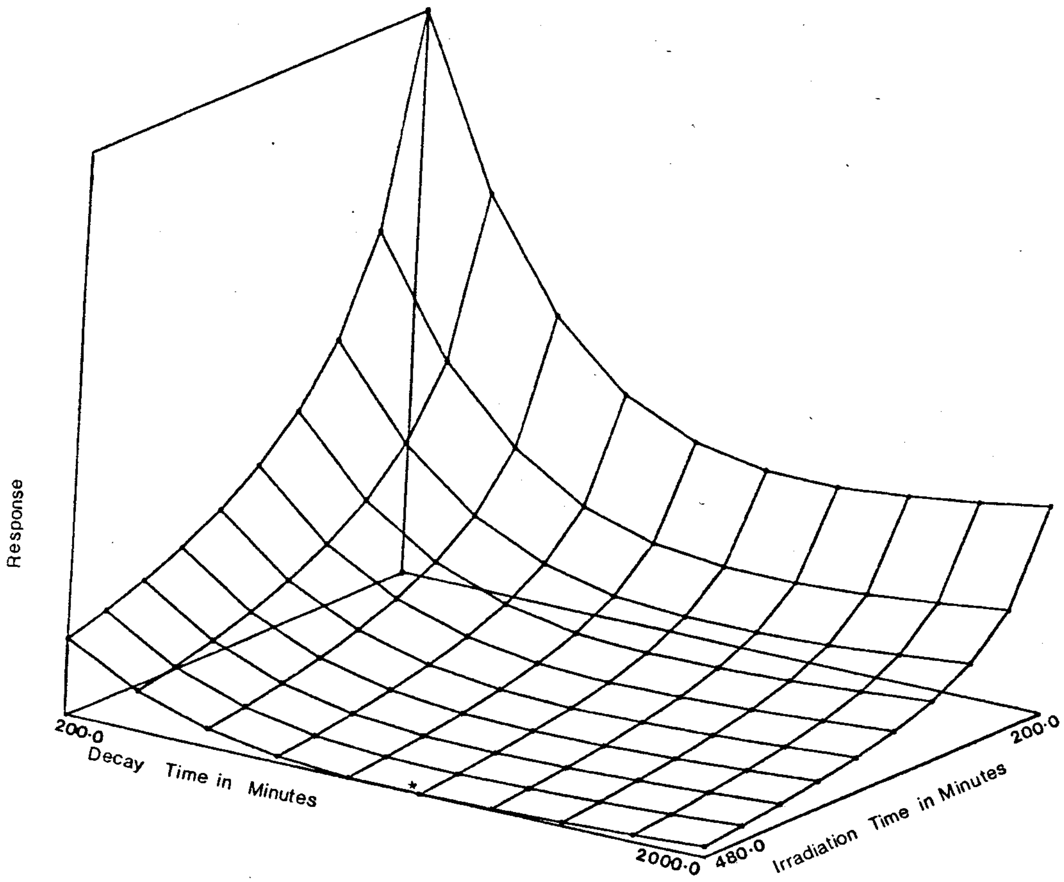


Table 15b. Table of Initial and Final Simplexes of ^{52}V

Run #	Vertex #	TD in Min.	TR in Min.	Type of Simplex
1	1	4.0	20.0	Initial
	2	1.0	5.0	
	3	10.0	45.0	
2	1	3.0	8.0	Initial
	2	10.0	3.0	
	3	16.0	50.0	
3	1	3.0	300.0	Initial
	2	0.6	2.0	
	3	10.0	15.0	
1	1	0.2000	22.57	Final
	2	0.2001	23.02	
	3	0.2001	22.95	
2	1	0.2000	23.01	Final
	2	0.2001	22.93	
	3	0.2001	22.98	
3	1	0.2000	23.00	
	2	0.2001	22.98	Final
	3	0.2001	22.75	

Figure 12. Grid plot of the Response surface of ^{52}V (Tc = 5.0 Min.)

N.B. The point of Simplex convergence is not within the grid (TD = 0.2 min, TR = 22-91 min.). However it can be seen from the response surface that it must be near (TD = 0.2 min and TR = 20.0 min.).

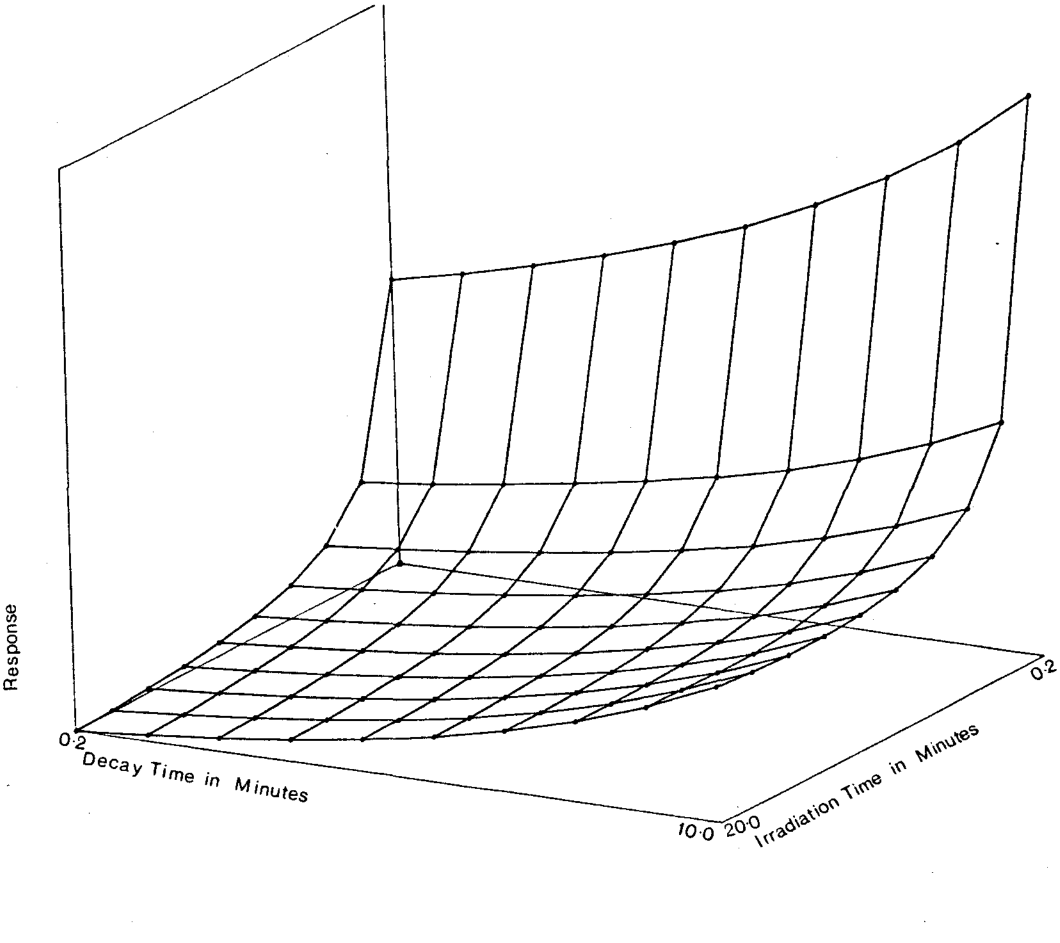
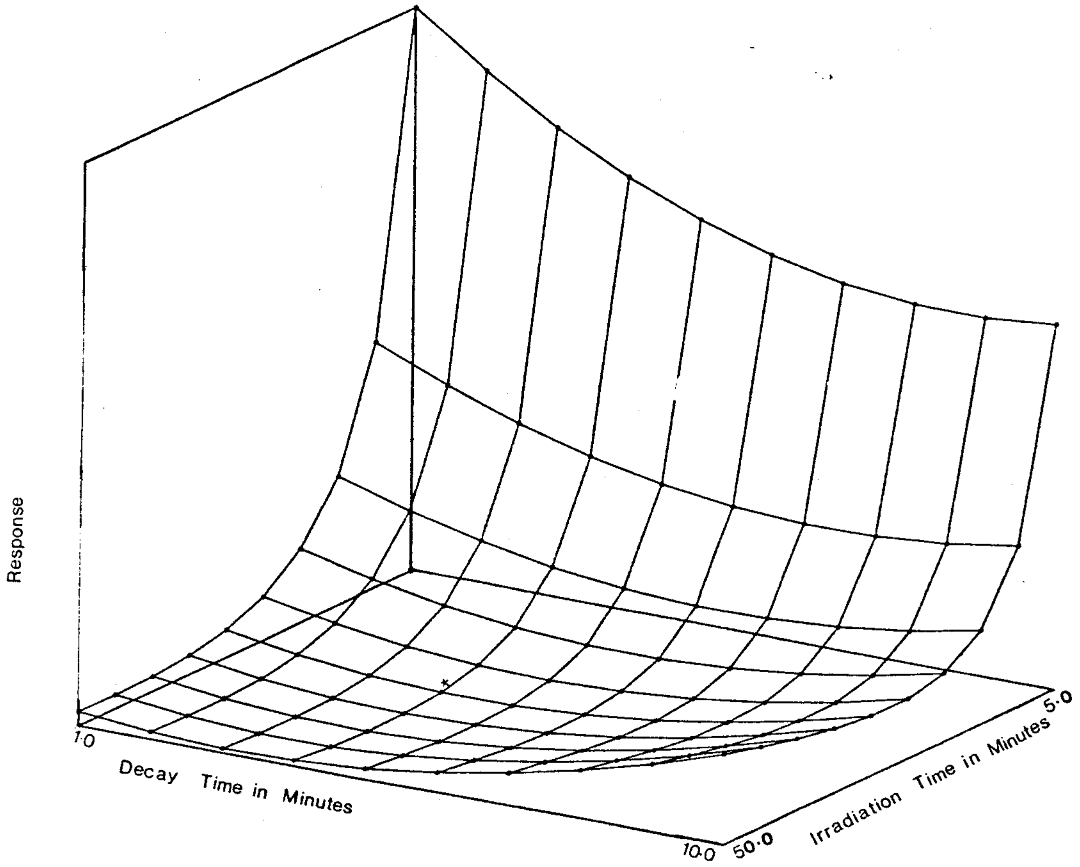


Table 15c Table of Initial and Final Simplexes of ^{27}Mg

Run #	Vertex #	TD in Min.	TR in Min.	Type of Simplex
1	1	1.0	70.0	Initial
	2	23.0	4.0	
	3	10.0	10.0	
2	1	10.0	20.0	Initial
	2	10.0	100.0	
	3	5.0	50.0	
3	1	20.0	60.0	Initial
	2	1.0	10.0	
	3	10.0	100.0	
1	1	3.902	37.82	Final
	2	3.853	37.66	
	3	3.908	37.39	
2	1	3.882	37.58	Final
	2	3.908	37.51	
	3	3.902	37.82	
3	1	3.891	37.60	Final
	2	3.878	37.83	
	3	3.902	37.77	

Figure 13. Grid Plot of the Response Surface of ^{27}Mg(Tc = 5.0).

* Point of Simplex convergence.



4.4 DISCUSSION OF RESULTS

(i) Detector Properties

The results for the detector properties as a function of energy are shown in Figures 7 to 10. In Figure 8, the point for the 511 Kev peak of Na-22 is furthest from the derived curve of the total detector efficiency. This point was not used to derive the function of total detector efficiency because of its distortion. The distortion was readily observed when using the Zimmerman method of finding the FWHM. Instead of a straight line curving towards the channel number axis at both ends of the line, the line obtained when the 511 Kev peak of Na-22 was linearized curved away from the channel number axis at the lower channel number end. The distortion of the peak shape is due to the high counting rate at which the Na-22 spectrum was acquired.

The definition of photopeak efficiency Pf(E) used to calculate the photopeak counts in this study is the product of the total detector efficiency d(E) and the photofraction f(E). If this photoefficiency is denoted as PFc, then

$$PFc = d(E)\cdot f(E)$$

Due to the approximations made in calculating some of the points used to derive the functional relationships of f(E) and d(E) to energy, the closeness of PFc to the photoefficiency function derived from fitting the results of Table 7 (i.e. the solid curve of Figure 7) was examined. The latter photoefficiency derived from fitting Table 7 data is denoted as PFe. The closeness of PFc to PFe was examined by comparing their values at a few selected energy values. Table 16 shows the results.

It is desirable that PFe and PFc give close results because this is one way of ensuring that spectra obtained from advance prediction agree with experimental spectra as long as spectral distortion is minimum. An alternative approach to ascertain that an advance predicted spectrum agrees with an experimental spectrum is to compare such spectra for a chosen sample. (It should be noted that advance predicted spectra in this study do not account for spectral distortion since this is not incorporated in the calculation of the spectrum. Disagreement of advance predicted spectrum with an experimental spectrum due to spectral distortion only. This can be taken care of by limiting the counting rate so that the distortion that would occur in the experimental spectrum is minimum. An alternative way of handling the disagreement would be to incorporate a calculational procedure to account for the distortion in the predicted spectrum.)

Table 16 Table of PFc and PFe

E (MeH)	f(E)	d(E) (%)	PFc (%)	PFe (%)
0.356	0.283	1.400	0.395	0.367
0.760	0.154	1.093	0.168	0.166
0.920	0.131	1.034	0.136	0.136
1.180	0.106	0.980	0.104	0.105
1.570	0.082	0.951	0.0785	0.0776
1.790	0.073	0.948	0.0695	0.0677

The apparent trend of the results of the comparison of PFe to PFe is that the difference between the two is more significant for energy regions where there are very few experimental data points of d(E) and f(E). The difference is more significant in the low energy region where both d(E) and f(E) vary most significantly with energy. This implies that more experimental points for d(E) and f(E) are required to obtain better agreement between PFc and PFe.

When considering the functional relationship of FWHM with energy, the most apparent feature is that the detector used in this study does not follow equation 32 which would require that the square of FWHM be a linear function of energy. This fact is acceptable since equation 32 is an imperical equation. An easy way to ascertain the reliability of data was to observe each peak's approximate FWHM in the MCA and compare it to the calculated value (in channel numbers). The calculated values when rounded off to an integer number always agreed with the number of channels observed on the MCA.

(ii) Optimum Times

The convergence regions of 3 initial simplexes for the elements shown in Table 12 were the sought optimum conditions. An average value of these times are shown in Table 14. The figure of merit used was a function proportional to the detection limit, that is, equation 23 in which the term accounting for room background is excluded. The peak base or Compton counts under the peak of interest was defined as

$$CBK = 3 \cdot FWHM \cdot CMP$$

A factor of 3 was used as in reference 41. Three examples confirming that the simplexes converge in the appropriate regions of detection limit from a mathematical point view of the search are shown in Figures 11 to 13.

From Table 14 it is seen that the optimum irradiation time is a few factors greater than the halflife (except for longlived isotopes in which the halflife is much greater than the maximum irradiation time - in this case the optimum irradiation time is the maximum irradiation time allowed). The optimum decay time for some shortlived isotopes ($\tau_{\frac{1}{2}} < 1$ hr) like ^{38}Cl in Table 14 seem rather unacceptable. From the experimental point of view one would tend to think that it is more reasonable that the decay time for ^{38}Cl be longer than predicted (especially after comparing it to that of ^{27}Mg) since the predicted decay time of 0.2 minutes leads to a high counting rate of ^{52}V and ^{28}Al which distort the acquired spectrum.

The disparity of the predicted decay time and the required experimental decay time for ^{38}Cl is due to the fact that a new factor; viz, counting rate has a significant effect. Since the program in its present form only takes account of the detection limit as a function of Compton interference, the predicted time is mathematically right in the current form of the program. This becomes easy to understand if we remember one point mentioned in Section 1.4; viz, there is an optimum decay time only if the Compton interfering gamma rays have a halflife less than half the halflife of the gamma ray of interest, otherwise the optimum decay time is zero (or in our case since a decay time of zero is not permitted we have 0.2 minutes which is the minimum decay time). For ^{38}Cl, there are no shortlived ray interferences hence the minimum decay time.

It should be mentioned that the effect of the appropriate counting rate is more serious than the above discussion implies for the short-lived isotopes. D. Burgess is currently modifying the program so that more reasonable times are predicted. There are other modifications necessary to the program as given in this thesis. Among the most obvious ones are the inclusion of the escape and 511 Kev peaks in the predicted spectrum when high energy gamma rays are considered. This requires the use of appropriate sources in calibrating the detector so that equations of escape and 511 Kev peak efficienceies are included in the program. It is also preferable to use more sources to derive the total detector efficiency and photofraction. One more modification necessary is the incorporation of a subroutine which flags overlapping peaks (see ref. 41) when such peaks are chosen for optimization. In this thesis, this problem was overcome by deliberately choosing peaks which do not overlap or they do overlap, as in ^{51}Ti and ^{51}Cr, then the contribution due to one element is negligible when one is interested in the other element due to big differences in halflife.

4.5 CONCLUSION

This study was carried out to try to find the optimum irradiation and decay time so that Compton interference is minimum and hence the detection limit is optimum. Unlike previous methods of irradiation and decay time optimization these times were obtained automatically and simultaneously for an element of interest. The results predicted for irradiation time were such that the sample was irradiated for lengths of time equal to a few factors of the halflife of the active nuclide used to optimize for the element of interest. Such times ensured sufficient activation of the target isotope. Some examples of the decay times predicted are for isotopes of As-76, Zn-69m, Br-82 and Cr-51. It is usual in INAA that elements with a halflife of about day in a complex sample matrix are counted after about a day and those of a halflife of about a month or greater are coutned after about a month (see the counting schedule in Ref. 29 and the optimum times for As-76 in Ref. 2). The results predicted in this study agree with those in the literature.

For some elements, the predicted decay times were not experimentally acceptable because decay times, in INAA are sometimes chosen to ensure acceptable counting rates and Compton interference during the counting interval. When these two requirements are mutually exclusive spectra acquired using the decay time predicted to have minimum Compton interference are distorted due to high counting rates. Calculation of the detection limit from such spectra is obviously unacceptable.

An example of this is Cl-38. The aspect of optimization incorporating the effects of counting rate on detection limit were not considered in this work and corrections necessary to achieve experimentally acceptable pairs of irradiation and decay time taking counting rate into account are being carried out by D.D. Burgess.

APPENDIX A

DETERMINATION OF PHOTOFRACTION AND TOTAL DETECTOR EFFICIENCY USING A NUCLIDE EMITTING TWO GAMMA RAYS (Ref. 33)

CASE 1. Total detector efficiency and photofraction assigned to weighted mean gamma line.

When determining total detector efficiency for monoenergetic gamma rays, the required parameters as shown by equation 9 are the total number of events detected and the total number of gamma rays emitted. When more than one gamma line is emitted by the source the total counts due to any one line cannot be easily determined without resorting to approximations because of overlapping of the detected events.

When the gamma lines are reasonably close, the assumption that the total detector efficiency and photofraction of a gamma energy lying between the two lines are approximately equal to the mean of detector efficiency and mean photofraction is reasonable as long as the intensities of the two peaks are equal. One can then readily use the mean parameters as required by equations 9 and 12 for the detector efficiency and photofraction. In the event that the two peaks do not have the same intensity, it is better to use a weighted mean gamma line and weighted mean parameters.

For example if the two lines are of energy E_1 and E_2, the weighted mean gamma line energy is:

$$Ea = (E_1 \cdot I_1 + E_2 \cdot I_2)/(I_1 + I_2) \qquad \text{A.1.}$$

where I_1 - is the intensity of E_1 and

I_2 - is the intensity of E_2.

The weighted mean area is:

$$Pa = (P_1 \cdot I_1 + P_2 \cdot I_2)/(I_1 + I_2) \qquad \text{A.2.}$$

where P_1 - is the peak area of the line with energy E_1 and

P_2 - is the peak area of the line with energy E_2.

The mean activity is:

$$Aa = (A_1 \cdot I_1 + A_2 \cdot I_2)/(I_1 + I_2) \qquad \text{A.3.}$$

where A_1 - is the activity of the line with energy E_1 and

A_2 - is the activity of the line with energy E_2.

From these values, equations 9 and 12 can be readily used to find the total detector efficiency and photofraction for the mean peak.

CASE 2. Decomposing the spectrum so that only parameters of one line are determined.

Decomposing a spectrum of two gamma rays relies on a procedure similar to splicing. This is used when two gamma lines are far apart since using the approximation of weighted mean is no longer valid. What is required is that one of the gamma rays lies in a region which has already been calibrated for photofraction. In such a case, since peak area determination is not a problem, equation 12 can be easily used to calculate the total number of events (i.e. Compton and photopeak events) due to the peak in the calibrated region. Subtraction of this calculated value from the integral of all detected events up to the peak lying outside the photofraction calibrated region gives the net count of this gamma line. The following equations make the explanation clearer.

Let E_1 and P_1 be the energy and peak count of the line in the calibrated region and let E_2 and P_2 be the energy and peak count whose detector efficiency and photofraction are sought. Let the net integral of counts up to the line whose parameters are sought be NP. If the photofraction of E_1 is f_1, then from equation 12, the total number of counts due to E_1 is:

$$NP_1 = P_1/f_1 \tag{A.4.}$$

So that the total number of counts due to E_2 is:

$$NP_2 = NP-NP_1 \tag{A.5.}$$

Once the net peak count of the line outside the calibrated region are known, equations 9 and 12 can then be used to find the total detector efficiency and photofraction (39).

APPENDIX B

DESCRIPTION OF THE PROGRAMS

1.1 PROGRAM SACOMP.

1.1.1 INPUT

IFN - name of file in which the arrays EN, ACTSAM, HTIME and AZAAPM are stored on a disk.

1.1.2 OUTPUT

EN	- array of all gamma ray energies emitted by the sample.
ACTSAM	- array of saturation activity for each emitted gamma ray per gram of sample.
HTIME	- array of halflife of every neutron activation produced nuclidic state in the sample entered for.
AZAAPM	- array of code identifying every active nuclidic state produced during activation.

1.1.3 DESCRIPTION

SACOMP.COM allows entry of the sample composition, the isotope parameters and the energies they emit. The program also calculates the saturation activity per gamma ray once the neutron flux is specified. Each emitted gamma ray, its saturation activity and halflife together with the code identifying the nuclidic state which emitted it are stored on a disk.

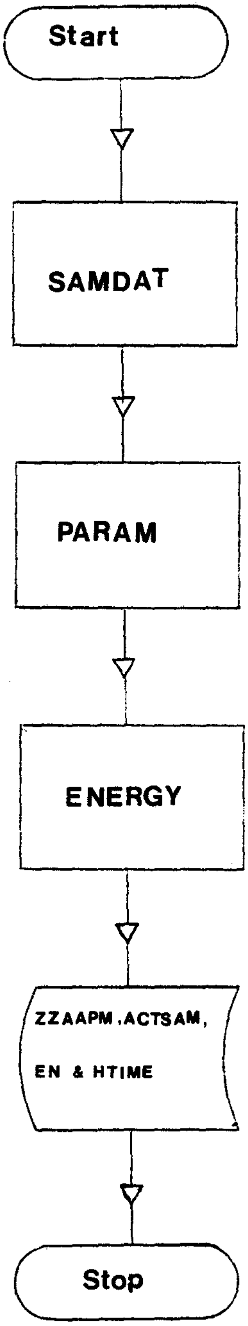
1.1.4 ROUTINES CALLED

SAMDAT

PARAM

ENERGY

Figure 14. Flowchart of program SACOMP



1.2 SUBROUTINE SAMDAT

1.2.1 INPUT

N - an integer specifying the number of elements in the sample.

Z - array of the atomic number of each element in the sample.

AT - array of the atomic weight of each element in the sample.

AC - array of % fraction of each element in the sample.

N1 - an integer allowing for the option to get a hardcopy of the entries so that they can be examined for mistakes which have to be corrected before returning to the SACOMP.

1.2.2 OUTPUT

Z, AT and AC when N1 specifies a hardcopy.

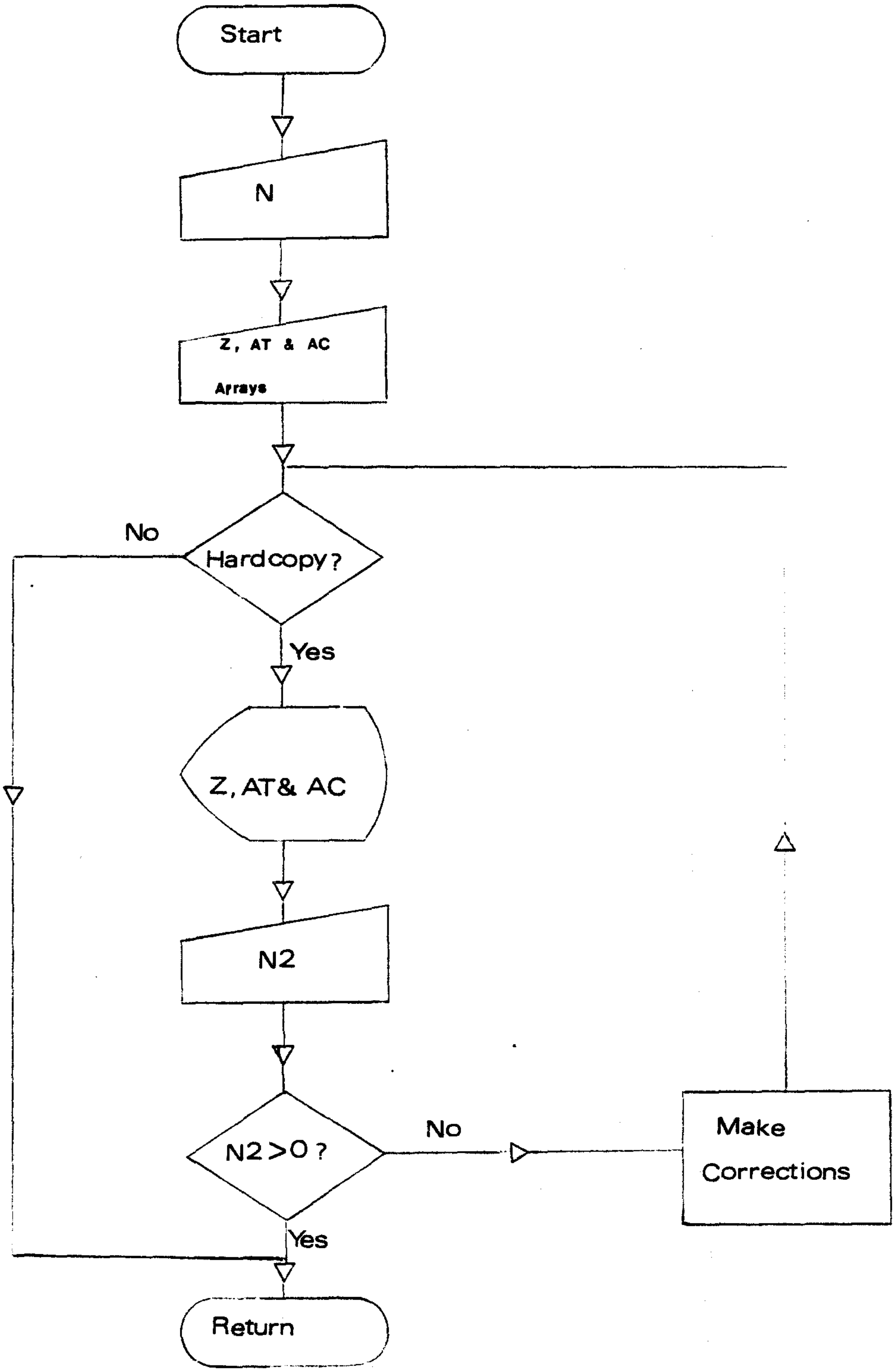
1.2.3 DESCRIPTION

SAMDAT enters the sample composition once the number of elements have been specified. It then prompts the user to obtain a hardcopy of the entries so that they can be examined for mistakes which have to be corrected before returning to the SACOMP.

1.2.4 ROUTINES CALLED

None.

Figure 15. Flowchart of subroutine SAMDAT



1.3 SUBROUTINE PARAM

1.3.1 INPUT

K - an integer specifying the number of radioactive nuclidic states produced during activation of each element.

ZZAAPM - an array of real numbers used to specify radioactive nuclidic states. The construction of ZZAAPM is as follows:

ZZ - is atomic number, AAP - is the mass number of the radioactive nuclide and M specifies whether the nuclide is in its ground state or is meta. M can take values from 0 to 9; by convention one specifies ground state nuclides by using the same number e.g. 9; ZZAAPM must have 6 digits.

XS - an array of reaction cross section in barns leading to a gamma emitting nuclidic state

HL - an array of halflife of gamma emitting nuclidic states. HL is in minutes.

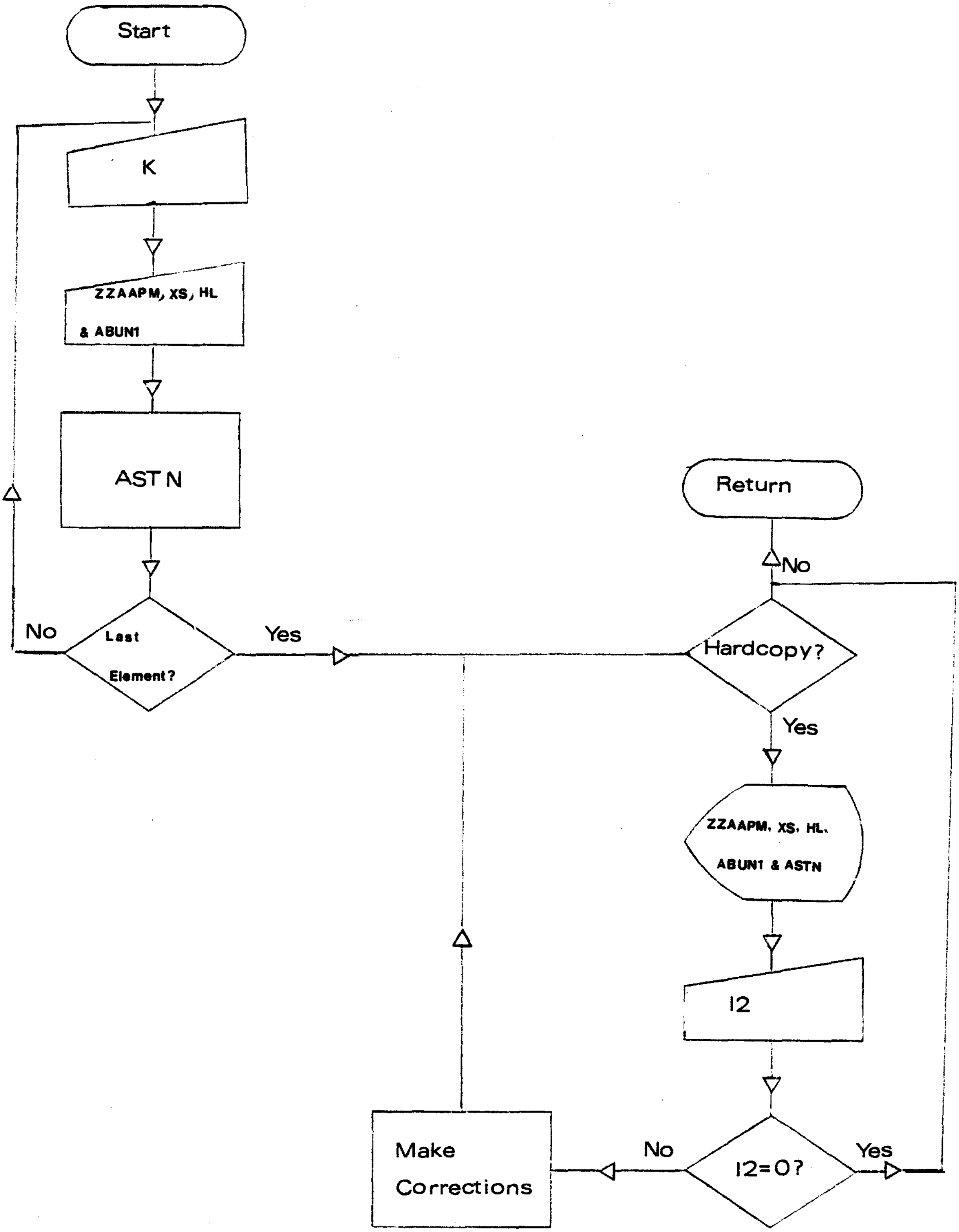
ABUN1 - an array of % isotopic abundance of the target isotope leading to a nuclidic state.

I1 - an integer used to specify the option to obtain a hardcopy of the entries or not.

I2 - an integer specifying the total number of entries to be corrected.

I3 - an integer specifying ZZAAPM, XS, HL and ABUN1 to be corrected.

Figure 16. Flowchart of subroutine PARAM



1.3.2 OUTPUT

When the appropriate I1 is specified, a hardcopy of ZZAAPM, XS, HL, ABUN1 and ASTN can be obtained and examined for any mistakes.

1.3.3 DESCRIPTION

The subroutine specifies an atomic number in the sample and requests for the number of radioactive nuclidic states which can be produced for each element. It then prompts the user to enter the code (sometimes referred to as product code) of each nuclidic state of the element, its halflife, the reaction cross section of the target isotope and the % isotopic abundance leading to this nuclidic state. The user can then obtain a hardcopy of the entries to check for any mistakes so that they can be corrected. The subroutine also calculates the saturation activity of each isotope per gram of sample. This saturation activity is stored in the array ASTN.

1.3.4 ROUTINES CALLED

None

1.4 SUBROUTINE ENERGY

1.4.1 INPUT

K - an integer specifying the number of significant gamma rays emitted by each nuclidic state (which can occur above the discriminator channel ≈ 100 Kev).

EN - array of gamma rays emitted by radioactive nuclidic states. The energies are entered in Kev (> 100 Kev).

ABUN2 - array of % emission intensity of each emitted gamma energy (> 1%).

N1 - an integer specifying option to obtain a hardcopy of the entries.

N2 - an integer indicating number of entries to be corrected.

N3 - an integer specifying the entries to be corrected.

1.4.2 OUTPUT

When N1 is chosen appropriately, a hardcopy of EN, ABUN2 and ACTSAM can be obtained. Necessary correction to EN and/or ABUN2 can be carried out after examining the hardcopy.

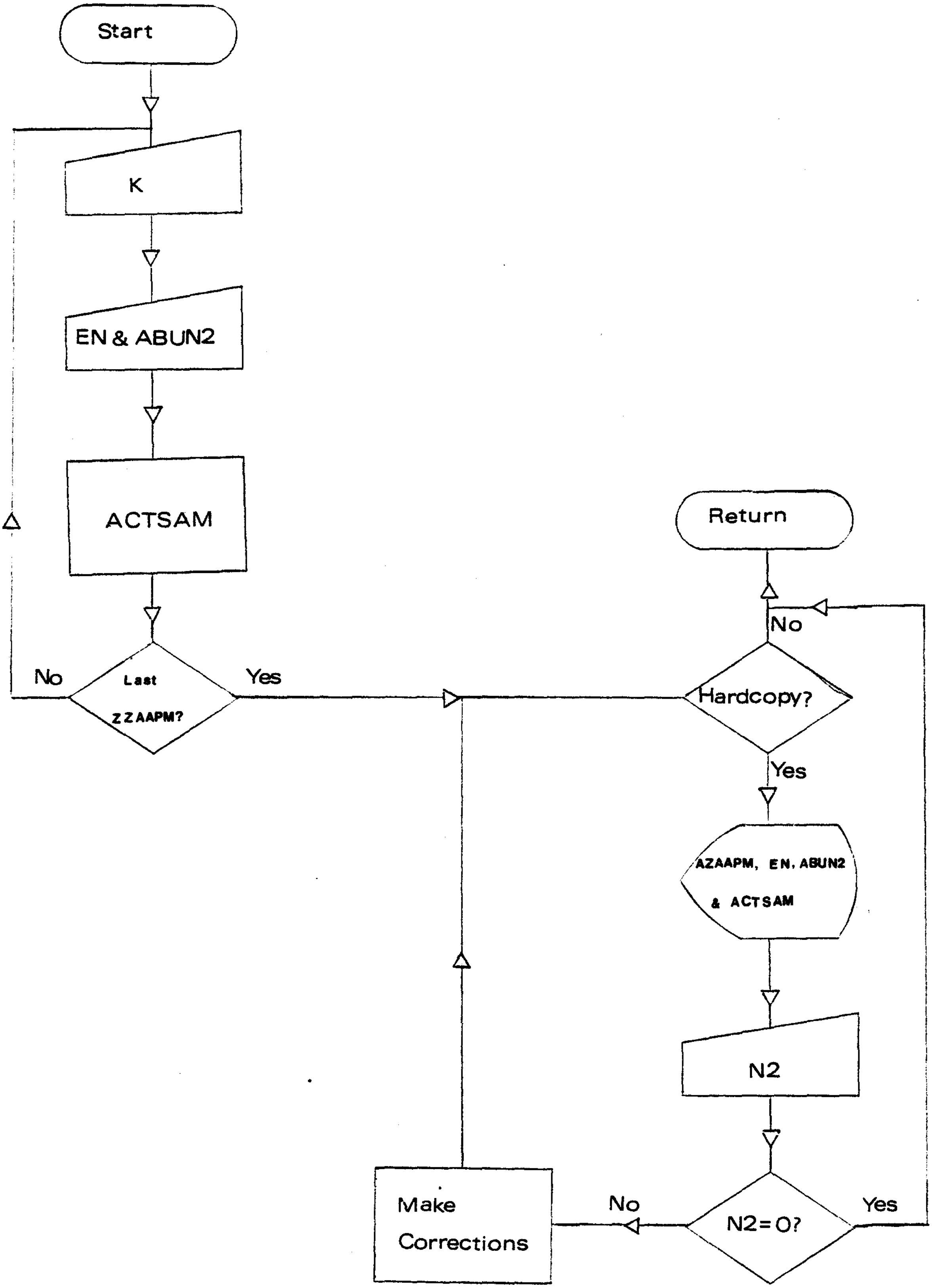
1.4.3 DESCRIPTION

The subroutine prompts one to enter the number of gamma rays emitted by each nuclidic state; one then enters these energies and their emission intensities. The subroutine then calculates the saturation activity for each gamma ray (array ACTSAM). A hardcopy of the entries can be obtained for examination of entries. Correction of the mistakes in the entries is the final step performed before returning to SACOMP.

1.4.4 ROUTINES CALLED

None

Figure 17. Flowchart of subroutine ENERGY



2.1 PROGRAM SIMPLEX

2.1.1 INPUT

HALT4 - a real number to which the standard deviation of the response at the simplex vertices (STDEV)(R), is compared to. If STDEV(R) < HALT4, the simplex is halted.

2.1.2 OUTPUT

Coordinates of the initial and last simplex.

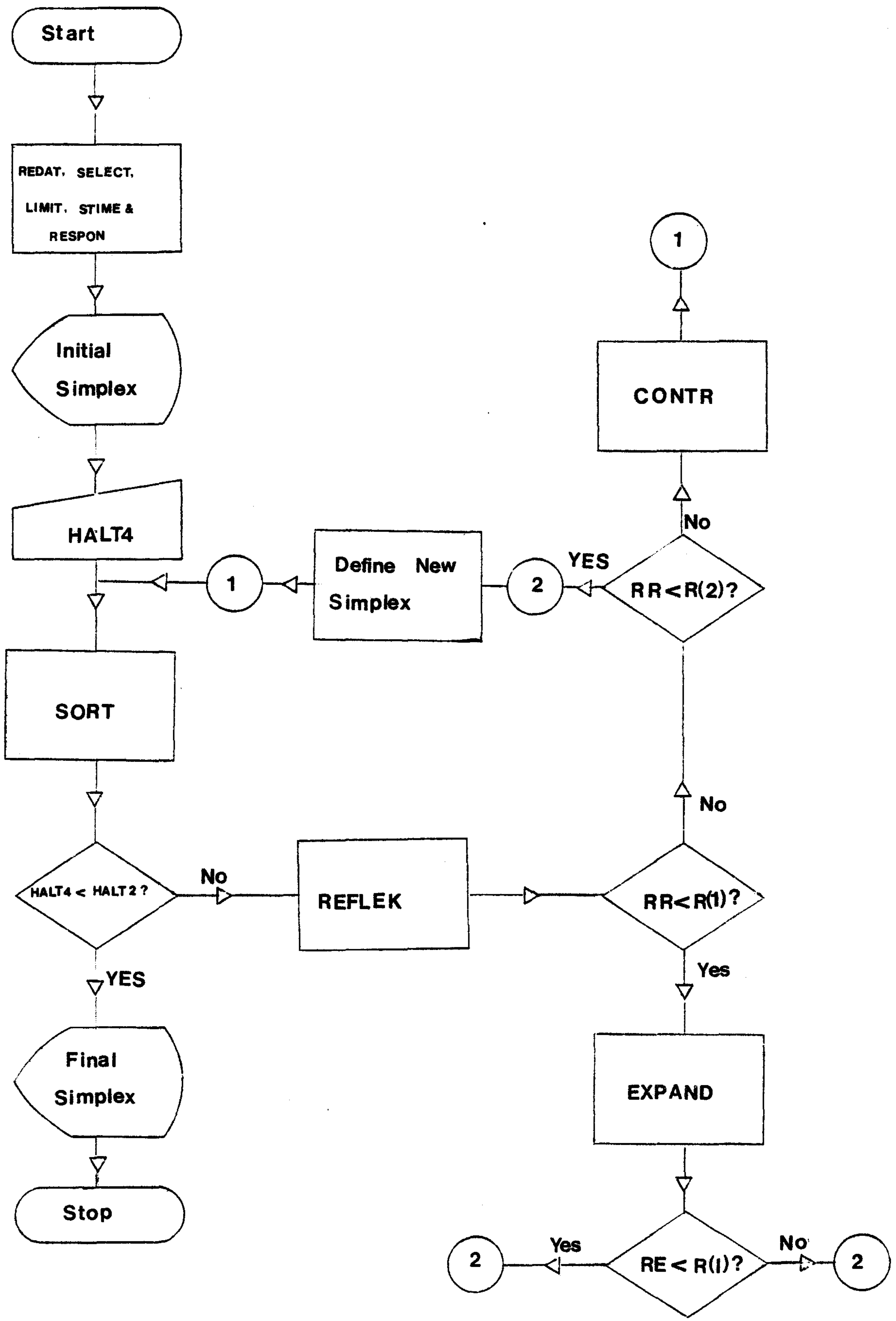
2.1.3 DESCRIPTION

The program accesses several subroutines necessary to perform. advance prediction of the spectrum of the sample. After the advance prediction, the detection limit is evaluated and used in the simplex translation routines until the convergence criteria is met.

2.1.4 ROUTINES CALLED

REDAT
SELECT
LIMIT
STIME
RESPON
SORT
REFLEK
EXPAND
CONTR
STDEV

Figure 18. Flowchart of program SIMPLEX



2.2 SUBROUTINE REDAT

2.2.1 INPUT

IFN - the filename in which the arrays EN, ACTSAM, HTIME and ZZAAPM (or AZAAPM) were stored on the disk.

EN - same as in SACOMP.COM

ACTSAM - same as in SACOMP.COM

HTIME - same as in SACOMP.COM

ZZAAPM - same as in SACOMP.COM

L - number of gamma rays in the spectrum.

2.2.2 OUTPUT

None

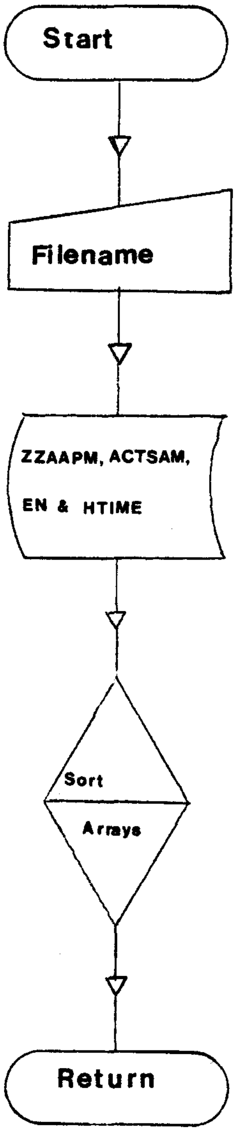
2.2.3 DESCRIPTION

The subroutine reads the data file made in SACOMP.COM thus entering the arrays EN, ACTSAM, HTIME and ZZAAPM. These arrays are sorted in decreasing order of gamma energy for an easy Compton cumulative build up in SUBROUTINE SPEC. After sorting,the subroutine exits to the main program SIMPLEX.

2.2.4 ROUTINES CALLED

None

Figure 19. Flowchart of subroutine REDAT



2.3 SUBROUTINE SELECT

2.3.1 INPUT

IZ - an integer specifying the atomic number of the element to optimize for.

II - an integer specifying the gamma energy entry number of the element to be used for optimization.

N - integer used to make another choice of IZ in case of a mistake.

M - integer used to warn user that the chosen IZ does not exist in the sample composition.

JX - integer used to select physical unit (monitor or printer) on which to show all gamma rays from the chosen element for a convenient choice of II.

2.3.2 OUTPUT

I - entry number of gamma ray from element with atomic number IZ. (N.B. One chooses II from one of the I's.)

EN(I) - gamma ray energy

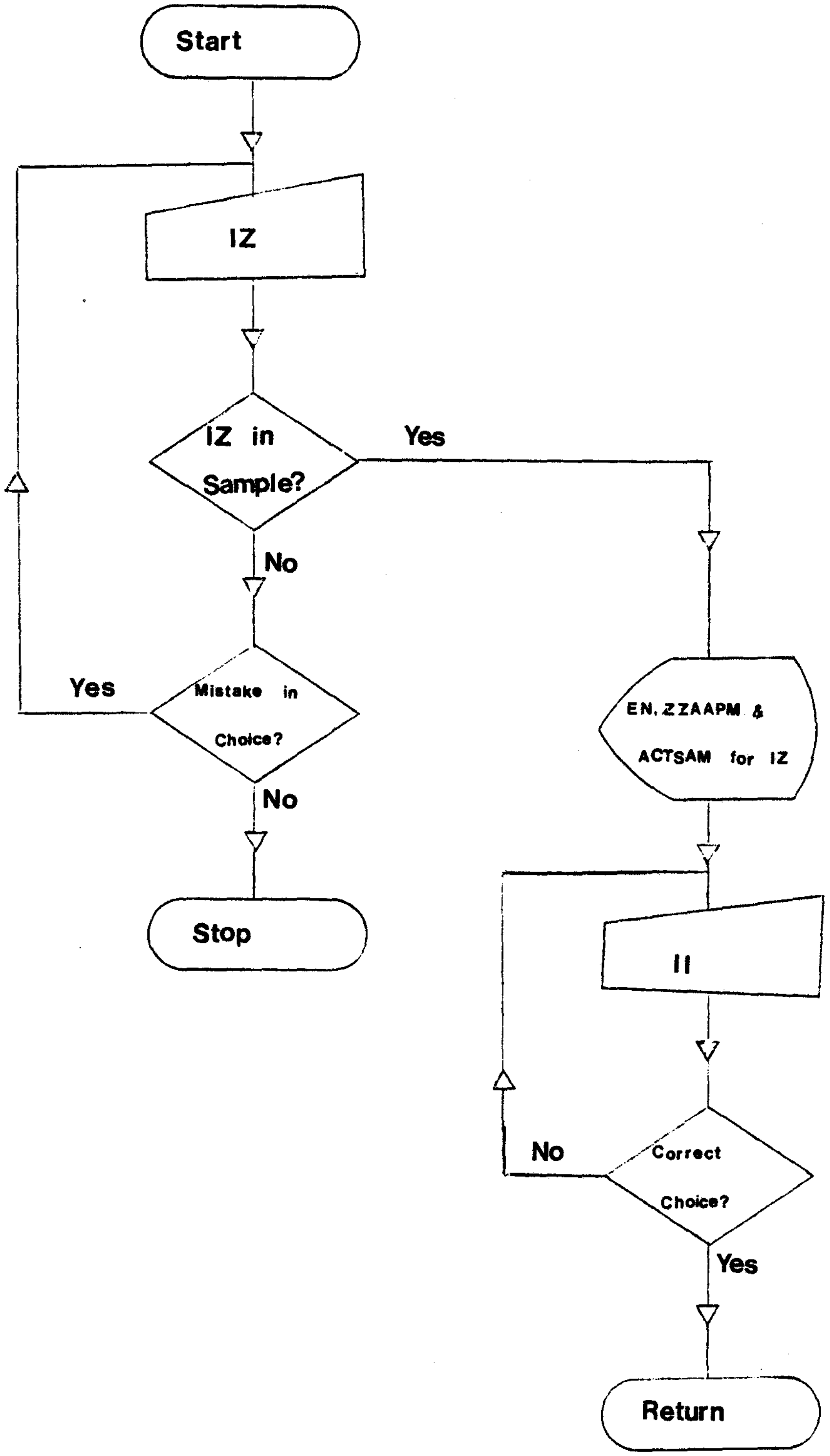
ZZAAPM(I)- code for the nuclidic state chosen.

ACTSAM(I)- saturation activity for gamma ray number I

2.3.3 DESCRIPTION

The subroutine is used to select the element to optimize for. After choosing a particular element by specifying its atomic number, the user is shown all the gamma rays from the element in decreasing order of gamma energy. By selecting a particular gamma energy entry number the user specifies the isotope,

Figure 20. Flowchart of subroutine SELECT



nuclidic state and gamma energy to optimize for the element of interest. After making the proper choice, the subroutine returns to SIMPLEX.

2.3.4 ROUTINES CALLED

None

2.4 SUBROUTINE LIMIT

2.4.1 INPUT

TDMIN - the minimum decay time allowed for the simplex.

TDMAX - the maximum decay time allowed for the simplex.

TRMIN - the minimum irradiation time allowed for the simplex.

TRMAX - the maximum irradiation time allowed for the simplex.

Tc - the decay time to be used in the advance prediction of the sample spectrum.

SMP - sample size in grams. This is chosen to give reasonable counting rate if possible.

N - integer giving option to change the limiting decay and irradiation times as well as the counting time.

N1 - integer giving option to change the sample size.

2.4.2 OUTPUT

None

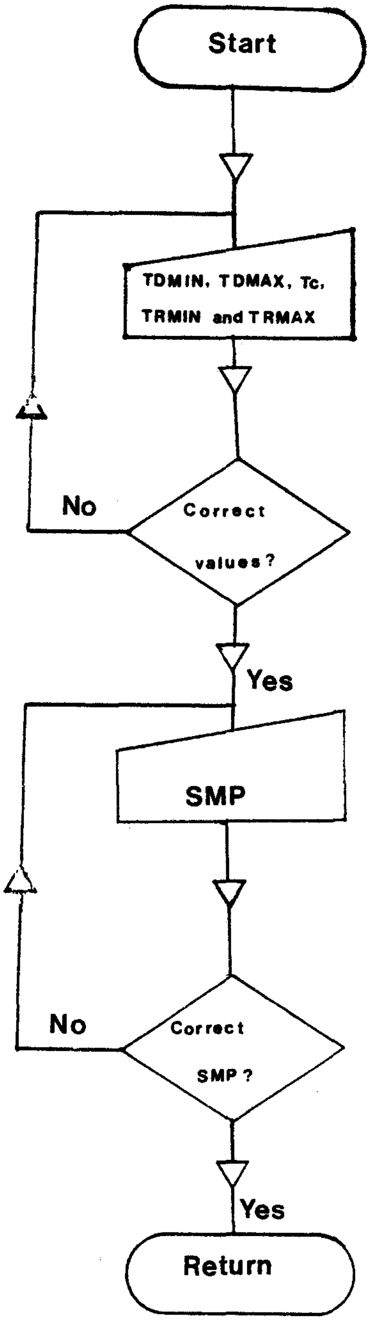
2.4.3 DESCRIPTION

The subroutine enters the boundary conditions of the simplex well as the counting time and sample size to be used in advance prediction of the spectrum.

2.4.4 ROUTINE CALLED

None

Figure 21. Flowchart of subroutine LIMIT



2.5 SUBROUTINE STIME

2.5.1 INPUT

TD - array of decay time of the initial simplex.

TR - array of irradiation time of the initial simplex.

N - integer giving an option to change the vertices of the simplex.

2.5.2 OUTPUT

TD - same as above.

TR - same as above.

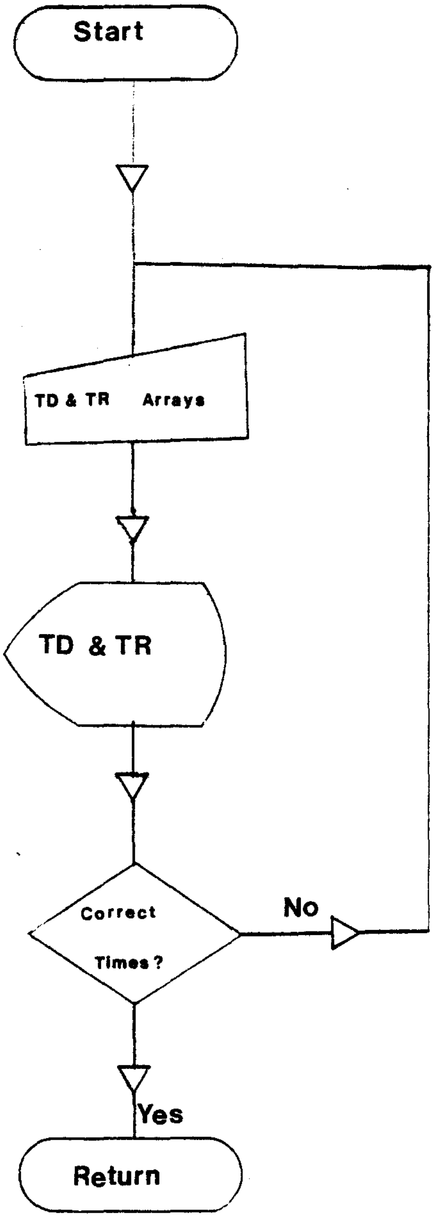
2.5.3 DESCRIPTION

The subroutine prompts the user to enter three sets of irradiation and decay time to define the initial simplex. The entries are displayed on the monitor. The user can change the vertices by an appropriate choice of N.

2.5.4 ROUTINES CALLED

None

Figure 22. Flowchart of subroutine STIME



2.6 SUBROUTINE RESPON

2.6.1 INPUT

2.6.2 OUTPUT

AAZZPM(II) - the nuclidic state being optimized for.

EN(II) - gamma energy used for optimization.

SMP - sample size.

Tc - counting time.

HTIME(II) - halflife of the nuclidic state giving the gamma rays.

II - entry number of the gamma energy used in the optimization.

PKC(II) - photopeak counts of the gamma energy used in the optimization.

I - vertix number.

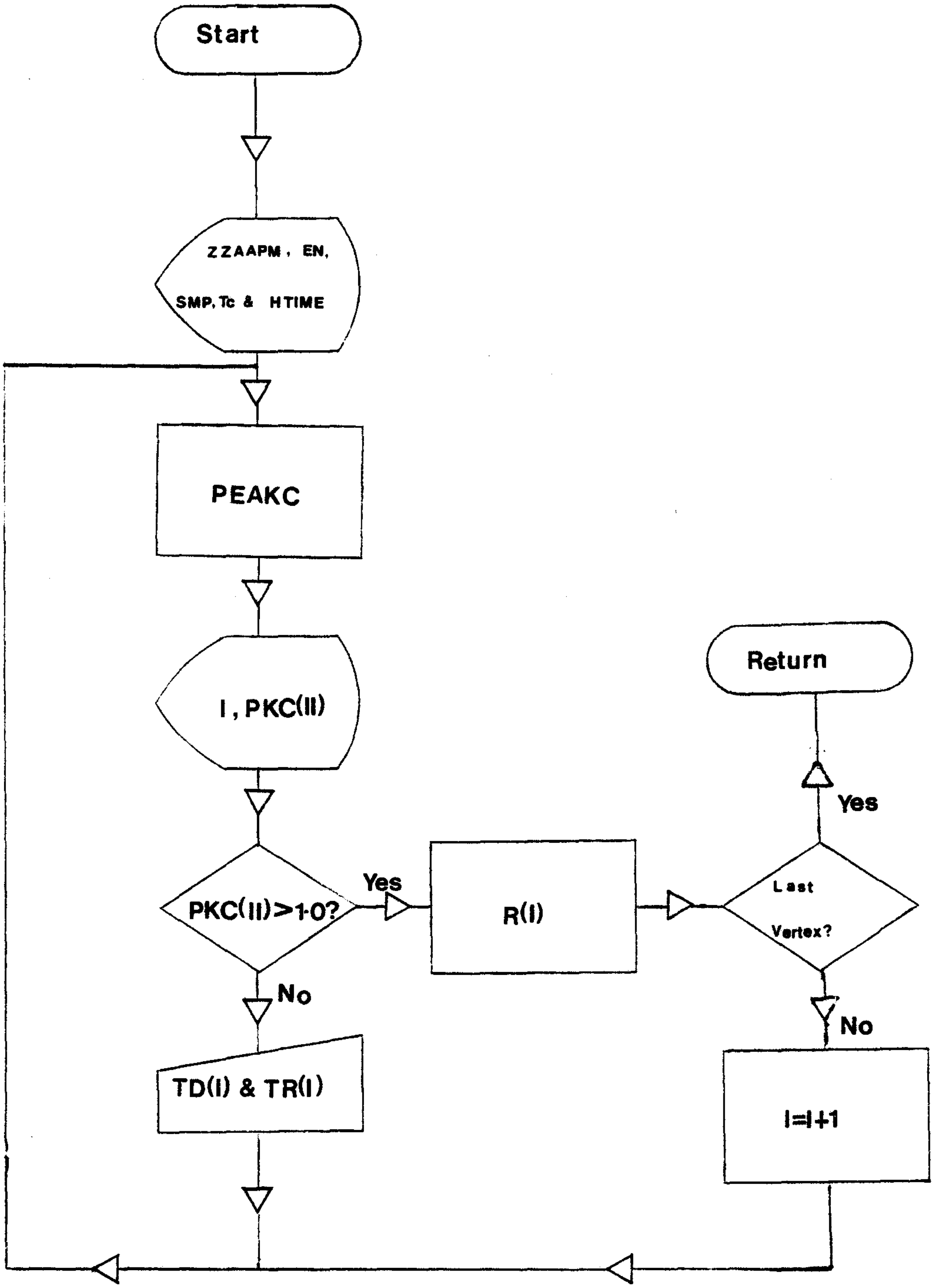
TD(I) - decay time for vertex number I in the initial simplex.

TR(I) - irradiation time for vertex number I in the initial simplex.

2.6.3 DESCRIPTION

The subroutine indicates the gamma energy being optimized for, the product radionuclide giving it and the halflife of this nuclide. The subroutine uses the advance prediction subroutines (SUBROUTINE PEAKC AND SPEC) to calculate the sample's spectrum for the initial simplex vertex and shows the user the peak counts for the peak being optimized at each vertex. If the peak count

Figure 23. Flowchart of subroutine RESPON



is less than one the subroutine prompts the user to reject this region since it seems that there are no peak counts registered. The user is prompted to try another set of times to define another prospective vertex.

If the peak counts are greater than one, then Compton background is calculated and the detection limit for each vertex is evaluated before returning to SIMPLEX.

2.6.4 ROUTINES CALLED

PEAKC

2.7 SUBROUTINE REFLEX

2.7.1 INPUT

None

2.7.2 OUTPUT

None

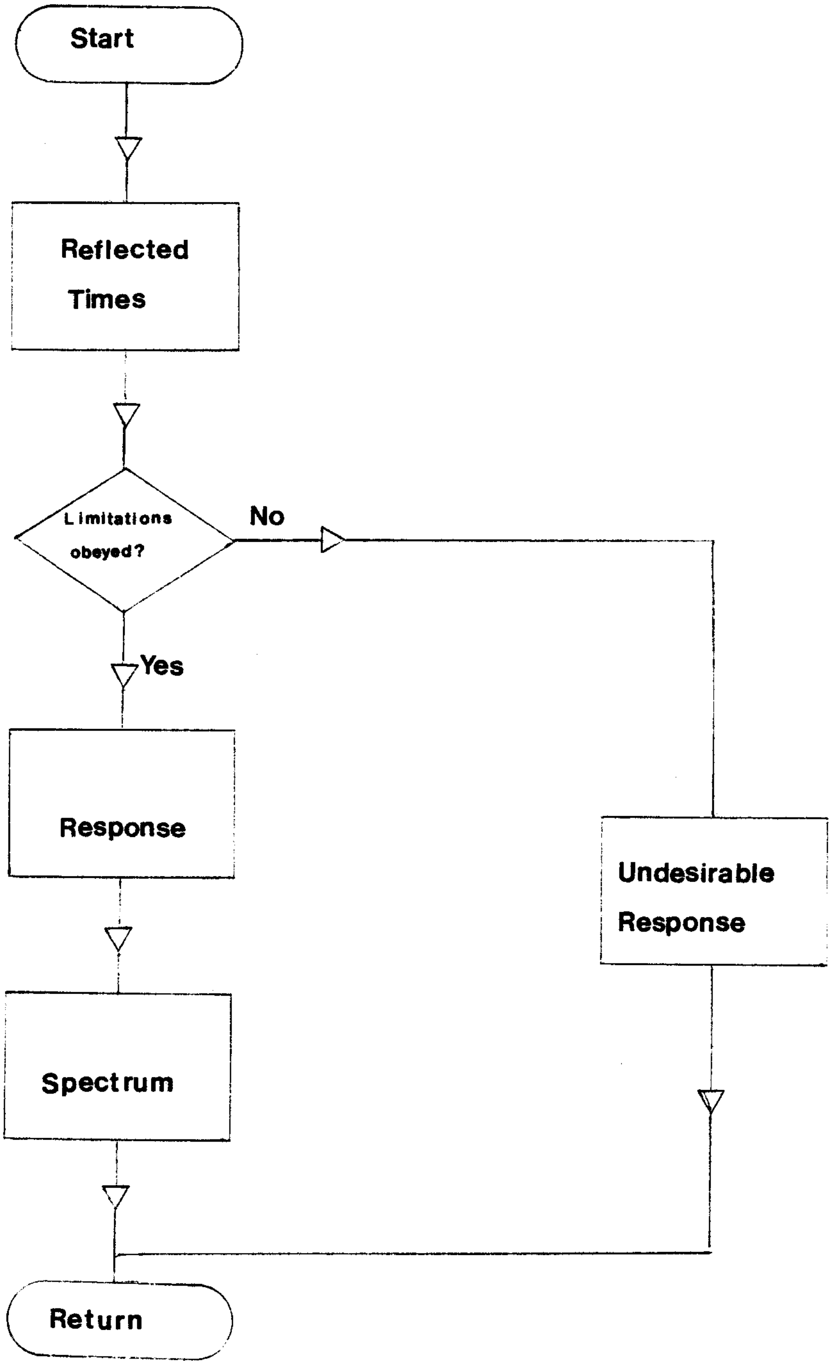
2.7.3 DESCRIPTION

The program evaluates the coordinates of the point at the centroid of the line joining the point with the lowest detection limit and the point with the next-to-lowest detection limit. This point is used to perform the reflection of the worst point. The resulting pair of decay and irradiation time are checked to see if they lie outside the search region. If one of the times lies outside the search region, the reflected point is assigned a very undesirable response and the subroutine returns to the main program. If the reflected point is within the search region, then advance prediction routines are called to obtain the spectrum at the reflected point. After calculating the spectrum, the subroutine returns to SIMPLEX which evaluates the reflected point according to the simplex move rules.

2.7.4 ROUTINES CALLED

PEAK

Figure 24. Flowchart of subroutine REFLEK



2.8 SUBROUTINE EXPAND

2.8.1 INPUT

None

2.8.2 OUTPUT

None

2.8.3 DESCRIPTION

When the reflected point's detection limit is lower than that for best point in the simplex the main program calls subroutine expand. The subroutine finds the times for the prospective expanded point. When these times define a point outside the search region the point is assigned a very undersirable response. The subroutine then submits control to the main program. If the point is within the search region the advance prediction routines are used to determine the sample's spectrum. The subroutine then returns to the main program which proceeds to find the detection limit which is compared to the current best point of the simplex.

2.8.4 ROUTINES CALLED

PEAKC

Figure 25. Flowchart of subroutine EXPAND

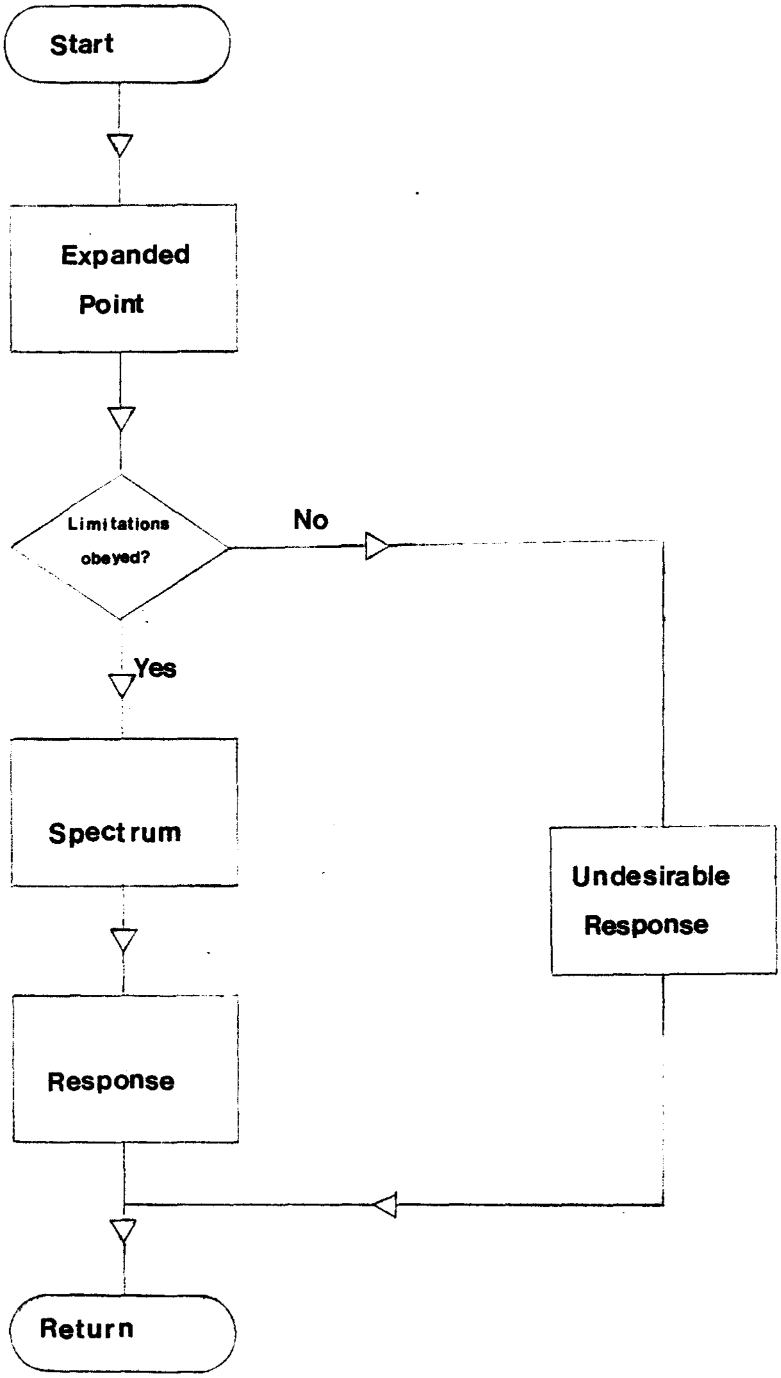
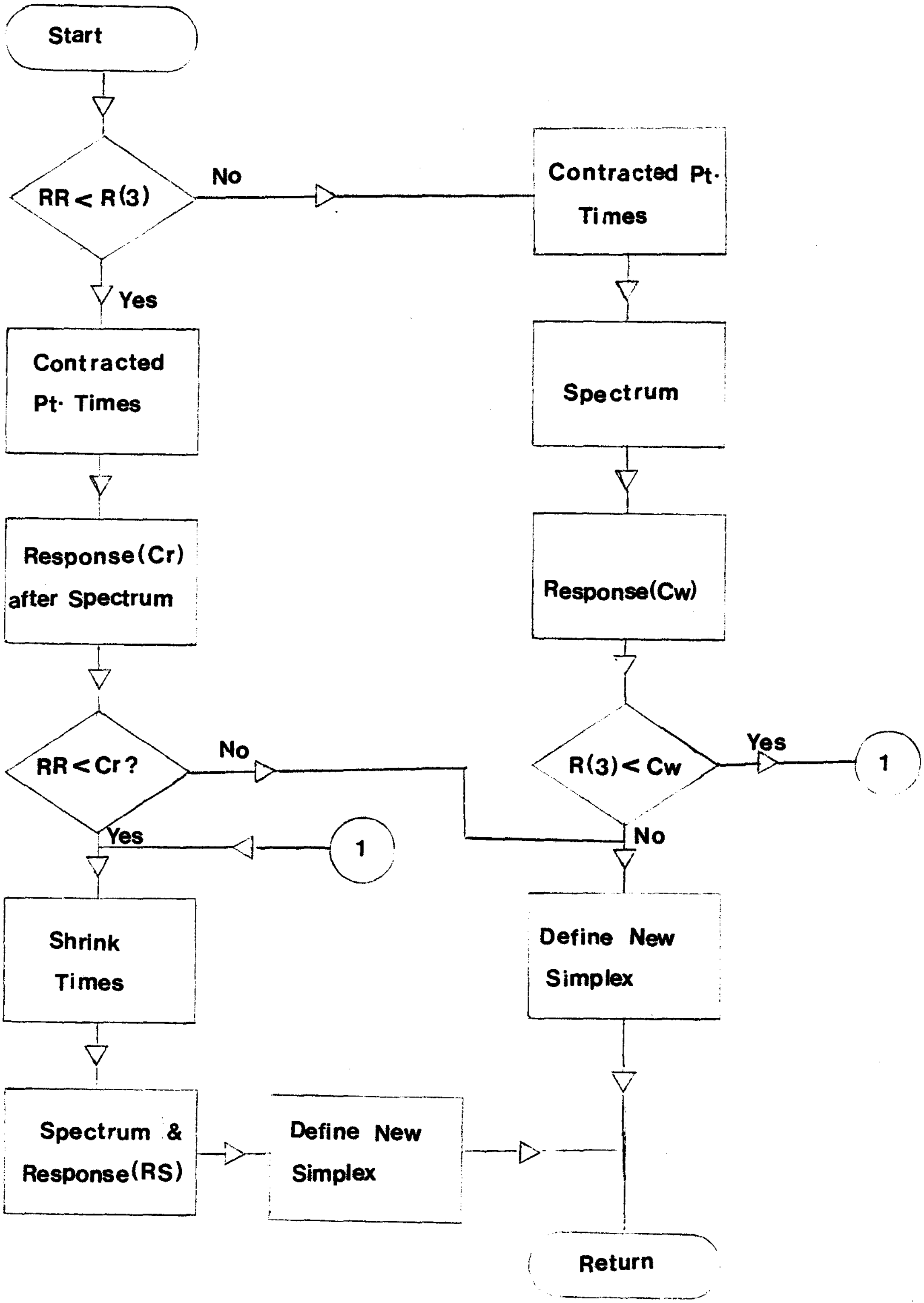


Figure 26. Flowchart of subroutine CONTR



2.9 SUBROUTINE CONTR

2.9.1 INPUT

None

2.9.2 OUTPUT

None

2.9.3 DESCRIPTION

When the reflected point is worse than the second best point, this subroutine is called to contract the simplex. If the reflected point has a response lower than the worst vertex, then the simplex is contracted about the reflected point. The contracted point automatically lies within the search region. The resulting times are used in the advance prediction routines. After constructing the spectrum, the detection limit is evaluated within the subroutine and compared to the reflected point. If the detection limit at the contraction point is worse than that at the reflected point the simplex undergoes massive contraction.

If the reflected point is worse than the worst point the simplex is contracted about the worst point. The contracted point times are used in the advance prediction routines and the detection limit determined after spectrum construction. The detection limit of the contracted point is compared to the worst point's detection limit. If the worst point is better than that at the contracted point, then the simplex undergoes massive contraction before yielding control to the main program.

Whatever the case, a new simplex is completely defined in the contraction subroutine before control is relinquished to the main program.

2.9.4 ROUTINES CALLED

PEAKC

2.10 SUBROUTINE PEAKC

2.10.1 INPUT

None

2.10.2 OUTPUT

None

2.10.3 DESCRIPTION

This is one of the two major subroutine for spectrum prediction implemented in PROGRAM SIMPLEX or GRID. It first determines total detector efficiency for each gamma ray. Then the total counts for each gamma ray at saturation are determined. These total counts are corrected for the counts obtainable for the given irradiation time using the saturation factor. The resulting counts are further corrected by multiplying with the decay factor. Then the prediction factor which takes into account the counting time effect of the determined count is multiplied to the results. The resulting total counts are scaled by multiplying with the sample size and stored in the array PKC. The subroutine then passes control to SUBROUTINE SPEC which finds, the photopeak counts and the cumulative Compton distribution.

2.10.4 ROUTINES CALLED

SPEC

Figure 27. Flowchart of subroutine PEAKC

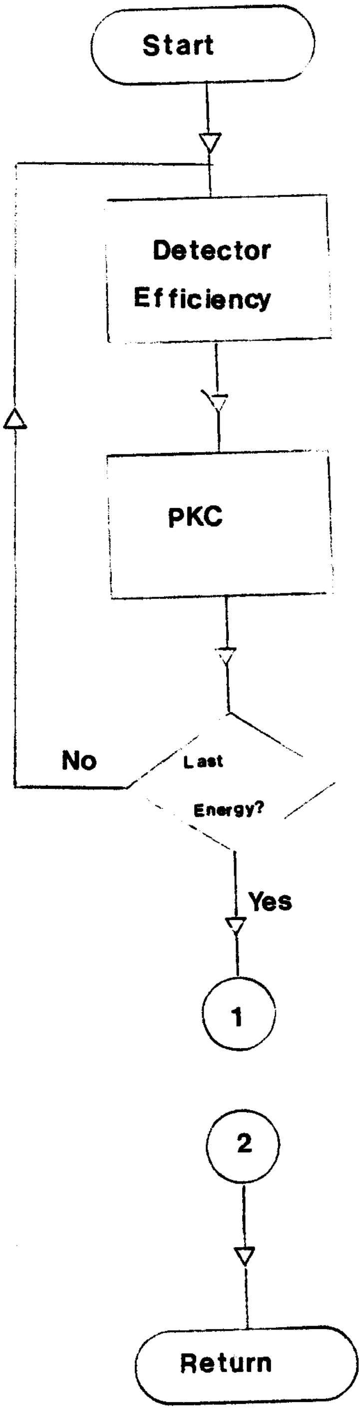
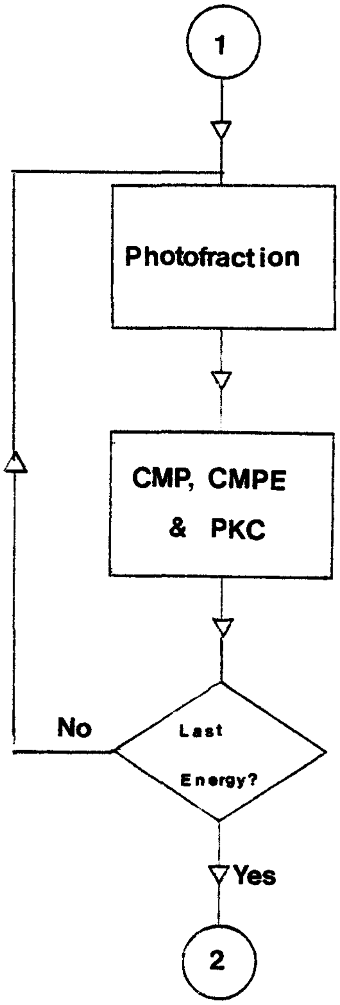


Figure 28. Flowchart of subroutine SPEC



2.11 SUBROUTINE SPEC

2.11.1 INPUT

None

2.11.2 OUTPUT

None

2.11.3 DESCRIPTION

This is the second major advance prediction routine. It starts by finding the photofraction of each gamma ray. Then the Compton fraction is readly determined from this. The product of the Compton fraction and the total counts from PEAKC (i.e. array elements in CMP corresponding to the gamma energy entries) gives the total Compton counts for each gamma ray energy. Then the Compton edge (CMPE) is determined which when divided into the total Compton counts gives the Compton distribution per Kev. The cumulative Compton background for the spectrum is built by adding the Compton contribution of all lower gamma energies to those of higher energies. After building the Compton background, the photopeak counts are determined by multiplying the photofraction with the total counts (PKC from PEAKC). This completes the prediction of the spectrum as performed in this program since the contribution of the escape and annihilation efficiencies were not determined for the detector.

For purposes of determining the detection limit of the element of interest, the Compton edge (CMPE(II) on which the peak of interest is superimposed upon is identified. The corresponding Compton counts in the Compton count array (CMP(II)) gives the Compton counts under the peak. After this identification, the subroutine returns to the PEAKC.

2.11.4 ROUTINES CALLED

None

2.12 FUNCTION STDEV(R)

2.12.1 INPUT

None

2.12.2 OUTPUT

None

2.12.3 DESCRIPTION

The function determines the standard deviation of the detection limits at the vertices of every simplex. The standard deviation is used to assess whether the simplex has converged.

2.12.4 ROUTINES CALLED

None

Figure 29. Flowchart of subroutine SORT

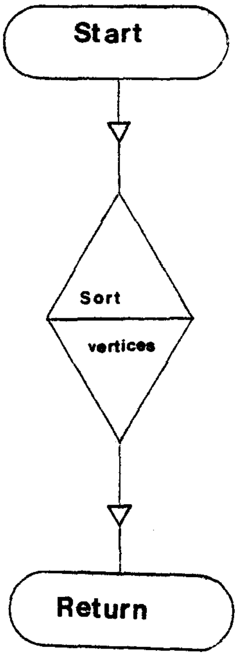
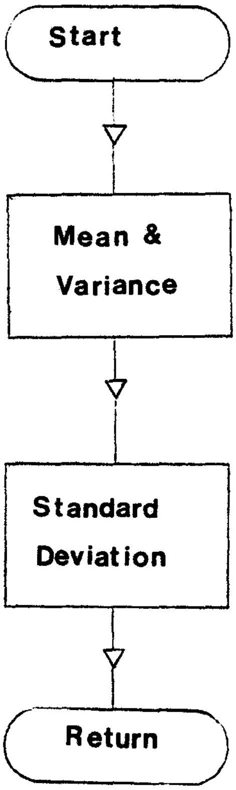


Figure 30. Flowchart of function STDEV



2.13 SUBROUTINE SORT

2.13.1 INPUT

None

2.13.2 OUTPUT

None

2.13.3 DESCRIPTION

The subroutine sorts the vertices of every new simplex into best, next-to-best and worst vertex so that logical decisions of simplex translation can be implemented.

2.13.4 ROUTINES CALLED

None

3.1 PROGRAM GRID

3.1.1 INPUT

N - an integer specifying the number of irradiation times in the grid.

N1 - an integer specifying the number of decay times in the grid.

IFN - filename in which grid data is stored on the disk.

3.1.2 OUTPUT

TD - array of decay time for the grid.

TR - array of irradiation time for the grid.

R - array of detection limit for the grid.

3.1.3 DESCRIPTION

The program uses the advance prediction routines to determine the detection limit of several pairs of irradiation and decay time. This set of coordinates TR(I), TD(I) and R(I) define a point in three dimensional space. (The arrays of these coordinates when plotted define detection limit topography.) The data of the arrays are stored on a disk.

3.1.4 ROUTINES CALLED

REDAT

SELECT

LIMIT

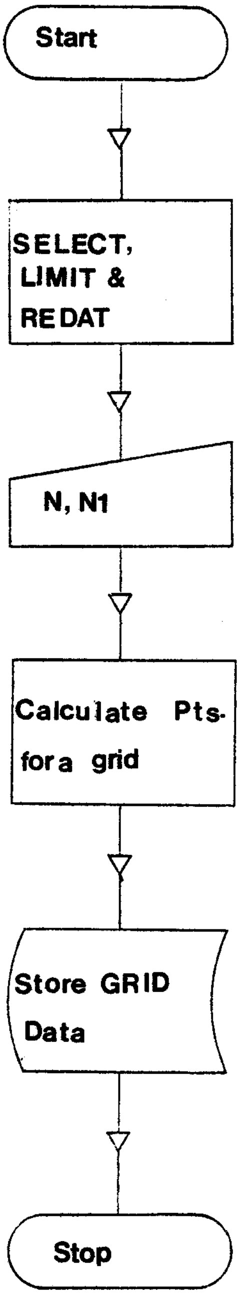
PEAKC

SPEC

3.1.5 COMMENTS

The subroutines above are the same as in the simplex program.

Figure 31. Flowchart of program GRID



However in LIMIT, the connotation of limiting times is that these are the boundaries of the grid. The plot of the GRID is obtained by using the Program PLOT3D (Ref. 42).

Appendix C1

Listing of Program SACOMP

```
      PROGRAM SACOMP
      BYTE IFN(14)
      COMMON/C1/EN(300), ABUN2(300), AZAAPM(300), ASTN1(300),
     +ACTSAM(300), HTIME(300)/C2/ZZAAPM(300), XS(300), ABUN1(300),
     +ASTN(300)/C3/AC(60), AT(60), Z(60)/C3/N,J,L
C
C     The program enters the sample composition, the isotope
C     parameters and their energies.
C
C
C     ROUTINES USED IN ORDER OF CALLING
C
C     SAMDAT - Enters Z, AT and AC
C     PARAM - Enters ZZAAPM, XS, HLTIME and ABUN1
C     ENERGY - Enters EN > 100 Kev and ABUN2 (usually > 1%)
C
C
C     DESCRIPTION OF ARRAYS:
C
C     EN                   array of gamma energies emitted (Kev.)
C     ABUN2                array of % emission intensities of gamma
C                          energies.
C     ZZAAPM & AZAAPM      code identifying the active nuclide or state
C                          in the sample; the letters stand for:
C          AZ or ZZ        atomic number;
C          AAP             mass number and
```

```
C          M                  is a number to specify the nuclidic state;
C                             for example, 9 could specify ground state.
C        ASTN & ASTN1         saturation activity per isotope per gram
C                             of sample.
C        ACTSAM               saturation activity per gamma energy per
C                             gram of sample.
C        HTIME & HL           halflives of the active isotopes or isomers
C                             (in minutes)
C        XS                   reaction cross-section of target isotope
C                             (in barns).
C        ABUN1                % isotopic abundance of target
C        AC                   sample composition (in %).
C        Z                    atomic number
C        AT                   atomic weight
C
C
C        DESCRIPTION OF OTHER PERTINENT VARIABLES:
C
C        Common Block C5 elements;
C
C          N                  number of elements known to be in sample.
C          J                  number of active isotopes and isomers in
C                             sample.
C          L                  number of gamma energies emitted by sample.
C
C        Start program execution
```

```
        CALL SAMDAT
        CALL PARAM
        CALL ENERGY
C
C       Write data onto disk.
C
        IF(L.NE.0) GO TO 15
        WRITE(1, 10)
 10     FORMAT('NO DATA CURRENTLY'/)
        STOP
 15     WRITE (1, 20)
 20     FORMAT ('ENTER FILENAME (CP/M STANDARD):')
        READ (1, 30)IFN
 30     FORMAT (40A1)
        CALL OPEN(6, IFN)
        DO 40 I=1, L
 40     WRITE (6, 50)EN(I), ACTSAM(I), HTIME(I), AZAAPM(I)
 50     FORMAT(4E14.7)
        ENDFILE 6
        STOP
        END

        SUBROUTINE SAMDAT
        COMMON/C3/AC(60),AT(60),Z(60)/C5/N,J,L
C
C       Enter the number of elements known to be in sample.
```

```
C
 101     WRITE(1, 10)
 10      FORMAT ('NUMBER OF ELEMENTS IN SAMPLE'/)
         READ(1, 20, ERR=101)N
 20      FORMAT(I3)
C
C        Enter sample composition.
C
         DO 7 I=1, N
 111     WRITE (1, 30)I
 30      FORMAT ('For element number', I3, 'please enter: '/1X, 'ATOMIC
        +NUMBER, ATOMIC WT. & % WT. FOR ELEMENT'/)
  7      READ(1, 40, ERR=111)Z(I), AT(I), AC(I)
 40      FORMAT(3E14.7)
C
C        Examine results to make corrections if necessary.
C
 121     WRITE (1, 50)
 50      FORMAT ('Do you want a hardcopy to examine entered data?'/X,'
        +1      YES,     2      NO'/)
         READ(1, 20, ERR=121)N1
         IF(N1.EQ.2)RETURN
         IF(N1.NE.1).AND.(N1.NE.2))GO TO 121
         WRITE(2, 60)
 60      FORMAT (1X, ' # ', 5X, 'ATOMIC #', 5X, 'ATOMIC WEIGHT', 5X,
        +'% WEIGHT/)
```

```
        DO 17 I=1, N
17      WRITE(2, 70) I,Z(I), AT(I), AC(I)
70      FORMAT(1X, 13, 5X, F4.0, 8X, F8.3, 7X, E12.5/)
131     WRITE(1, 80)
80      FORMAT('After examining hardcopy please enter: '/1x, '# OF
       +CORRECTIONS NEEDED'/)
        READ(1, 20, ERR=131)N2
        IF(N2.EQ.0)RETURN
        DO 27 I=1, N2
141     WRITE(1, 90)
90      FORMAT('ENTER THE ENTRY # TO BE CORRECTED'/1x,' (as on hard
       +copy)'/)
        READ(1, 20, ERR=141)N3
151     WRITE(1, 100)N3
100     FORMAT(1X, 'CORRECT ATOMIC #, ATOMIC WT. & % WT. FOR ENTRY'/1X,'
        #', I3/)
27      READ(1, 40, ERR=151)Z(N3), AT(N3), AC(N3)
        GO TO 121
        END


        SUBROUTINE PARAM
        COMMON/C2/ZZAAPM(300), XS(300), ABUN1(300), HL(300), ASTN(300)/
       +C3/AC(60), AT(60),Z(60)/C5/N,J,L
C
C       Initialise counter of active isotopes and isomers.
C
```

```
      J=0
      DO 7 I=1,N
C
C     Specify the number of gamma active nuclides and isomers for
C     each element.
C
 101  WRITE(1, 10)Z(I)
 10   FORMAT('For element with atomic #',F4.0,' enter: '1x, 'THE #
     +OF ACTIVE NUCLIDES & ISOMERS'/)
      READ(1, 20, ERR=101)K
 20   FORMAT(13)
C
C     Enter nuclear parameters for each element.
C
      DO 17 L1=1,K
      J=J+1
 111  WRITE(1, 20)Z(I),L1
 30   FORMAT('For atomic #',F4.0,' active nuclide or isomer', 13/1X,
     +'enter ZZAAPM, XS, HL and ABUN1'/)
      READ(1, 40, ERR=111)ZZAAPM(J), XS(J), HL(J), ABUN1(J)
 40   FORMAT(4E14.7)
C
C     Calculate the saturation activity of each isotope per gram of
C     sample.
C
 17   ASTN(J)=ABUN1(J)*XS(J)*6.023E7*AC(I)/AT(I)
 7    CONTINUE
```

```
C       Examine results and make corrections where necessary.
 121    WRITE(1, 50)
 50     FORMAT('Do you want a hardcopy to examine data?'/1X,'1
       +      YES;      2       NO'/)
        READ(1, 20, ERR=121)I1
        IF(I1.EQ.2) RETURN
        IF((I1.NE.1).AND.(I1.NE.2)) GO TO 121
        WRITE(2, 60)
 60     FORMAT(1X,' # ', 5X, 'ZZAAPM', 8X, 'RXN. XS.', 7X, 'HALFLIFE',
       +7X,' % ABUND.', 4X,' SAT. ACTIVITY/GRAM'/)
        WRITE(2, 70) (I, ZZAAPM(I),XS(I),HL(I),ABUN1(I),ASTN(I),I=1,J)
 70     FORMAT(1X, I3, 4X, F8.0, 4(3X, E12.5)/)
C
 131    WRITE(1, 80)
 80     FORMAT('After examining hardcopy enter: '/1X, '# OF CORRECTIONS
       +NEEDED'/)
        READ(1, 20, ERR=131)I2
        IF(I2.EQ.0) RETURN
        DO 27 I=1,I2
 141    WRITE(1, 90)
 90     FORMAT('ENTER THE ENTRY # TO BE CORRECTED'/1X,' (as on hardcopy'/)
        READ(1, 20, ERR=141)I3
 151    WRITE(1, 200)I3
 200    FORMAT('For entry #', I3/1X, 'ENTER THE CORRECT ZZAAPM, XS,
       +HL & ABUN1'/)
        READ(1, 40, ERR=151)ZZAAPM(I3), HL(I3), ABUN1(I3)
```

```
      AAT=FLOAT(INT(ZZAAPM(I3)/1.E4))
      DO 37  IP=1,N
37    IF(AAT.EQ.Z(IP)) ASTN(I3)=ABUN1(I3)*6.023E7*AC(IP)/AT(IP)
27    CONTINUE
      GO TO 121
      END
      SUBROUTINE ENERGY
      COMMON/C1/EN(300), ABUN2(300), AZAAPM(300), ASTN1(300), ACTSAM
     +(300), HTIME(300)/C2/ZZAAPM(300), XS(300), ABUN1(300), HL(300),
     +ASTN(300)/C5/N,J,L
C
C     Intialise energy counter.
C
      L=0
      DO 7 I=1,J
C
C     Specify the number of gamma energies emitted by each nuclidic
C     species.
C
101   WRITE(1, 10)ZZAAPM(I)
10    FORMAT('For a nuclidic species denoted as', F8.0/1X, 'ENTER THE
     +# OF GAMMA ENERGIES > 100. Kev'/)
      READ(1, 20, ERR=101)K
20    FORMAT(I3)
      DO 17 M=1, K
      L=L+1
```

```
 111      WRITE(1, 30)ZZAAPM(I),M
 30       FORMAT('For product code', F8.0/1X, 'ENTER:  ENERGY & %
         +EMISSION FOR GAMMA RAY #', I3/)
          READ(1, 40, ERR=111) EN(L), ABUN2(L)
 40       FORMAT(2E14.7)
          ASTN1(L)=ASTN(I)
          AZAAPM(L)=ZZAAPM(I)
C
C         Calculate the saturation activity for each gamma ray per gram of
          sample.
C
          ACTSAM(L)=ABUN2(L)*ASTN(I)*.01
 17       HTIME(L)=HL(I)
 7        CONTINUE
C
C         Examine entries and carry out corrections where necessary.
C
 121      WRITE(1, 50)
 50       FORMAT('Do you want a hardcopy to examine data?'/1X,' 1
         +       YES;       2       NO'/)
          READ(1, 20, ERR=121)N1
          IF(N1.EQ.2) RETURN
          IF((N1.NE.1).AND.(N1.NE.2)) TO TO 121
          WRITE(2,60)
 60       FORMAT(1X,' #', 5X, 'ENERGY', 5X, 'ABUNDANCE', 5X, 'ZZAAPM',
         +5X, 'SAT. ACTIVITY/GRAM'/)
```

```
        WRITE(2,70)(I,EN(I),ABUN2(I),AZAAPM(I),ACTSAM(I), I=1, L)
70      FORMAT(1X, I3, 4X, F7.2, 6X, F7.2, 2X, F8.0, 5X, E12.5/)
131     WRITE(1, 80)
80      FORMAT('After examining hardcopy:'/1X, 'ENTER # OF CORRECTIONS
       +NEEDED'/)
        READ(1, 20, ERR=131)N2
        IF(N2.EQ.0) RETURN
        DO 37 I=1, N2
141     WRITE(1, 90)
90      FORMAT(' ENTER ENTRY # TO BE CORRECTED'/1X,' (as on hardcopy)'/)
        READ(1, 20, ERR=141)N3
151     WRITE(1, 200)N3
200     FORMAT('CORRECT ENERGY & % ABUNDANCE FOR ENTRY #', I3/)
        READ(1, 40, ERR=151)EN(N3), ABUN2(N3)
        ACTSAM(N3)=ASTN1(N3)*ABUN2(N3)*.01
37      CONTINUE
        TO TO 121
        END
```

Appendix C2

Listing of Program SIMPLEX

```
      PROGRAM SIMPLX
      COMMON/B1/ACTSAM(300), HTIME(300),PKC(300),EN(300),CMP(300),CMP
     +E(300),ZZAAPM(300)/B2/TD(3),TR(3),PPKC(3),CC(3)/B3/TDMIN,
     +TDMAX,TRMIN,TRMAX,TC,SMP/B4/L,II,JJ/B5/HALT4
C
C       The program determines the optimum irradiation and decay time
C       for Neutron Activation Analysis.
C
C       ROUTINES USED.
C
C       REDAT
C       SELECT
C       LIMIT
C       STIME
C       RESPON
C       REFLEK
C       SORT
C       STDEV
C       EXPAND
C       CONTR
C       PEAKC
C       SPEC
C
C       DESCRIPTION OF SUBROUTINES
C
C       REDAT  - reads in the arrays EN, ACTSAM,HTIM & ZZAAPM from the
```

```
                 the data file made by SACOMP.
C       SELECT - enters the element and gamma ray to optimize for.
C       LIMIT  - enters the boundary times for the simplex search.
C       STIME  - enters the times for the initial simplex.
C       RESPON - calculates the spectra of the initial simplexes
C       SORT   - sorts the vertices of every simplex in increasing
C                order of the response.
C       REFLEK - reflects the worst point of every new simplex.
C       CONTR  - contracts the simplex.
C       EXPANDS- expands the simplex.
C       PEAKC  - calculates the total counts for every specified set
C                of irradiation, decay and counting time.
C       SPEC   - calculates the photopeak and Compton distribution
C                using data from PEAKC.
C       STDEV  - determines the standard deviation of the simplex
C                vertices.
C
C       COMMENTS:
C
C       Subroutine PEAKC is called whenever it is necessary to generate
C       a spectrum.  PEAKC always calls subroutine SPEC to complete the
C       spectrum generation.
C       The routines which call PEAKC are RESPON, REFLEK, EXPAND & CONTR.
C       Whenever a new simplex is defined, the vertices are sorted by
C       subroutine SORT.  After sorting their standard deviation is
C       calculated.  If this standard deviation falls below some pre-
```

```
C       selected value, simplex translation stops.
C
C       DESCRIPTION OF ARRAYS.
C
C       EN          - gamma array.
C       ACTSAM      - saturation activity for each gamma ray for a gram of
C                     sample.
C       ZZAAPM      - code identifying the nuclidic state emitting the
C                     corresponding gamma ray.
C       HLIFE       - the halflife of gamma emitting nuclidic state.
C       PKC         - total counts detected for a gamma ray in PEAKC.
C                   - photopeak counts for a gamma ray in SPEC.
C       CMP         - Compton counts per Kev.
C       CMPE        - Compton edge
C       R           - response function proportional to detection limit
C                     of current simplex.
C       TR          - irradiation times of current simplex
C       TD          - decay time of current simplex.
C       CC          - Compton counts under the peak of interest for the
C                     current simplex.
C       PPKC        - photopeak counts for the current simplex.
C
C       DESCRIPTION OF OTHER PERTINENT VARIABLES
C
C       SMP         - sample size in grams.
C       CMP(JJ)     - Compton counts under peak of interest.
```

```
C       PKC(II)  - peak of interest.
C       TRMIN    - minimum irradiation time.
C       TRMAX    - maximum irradiation time.
C       TDMIN    - minimum decay time.
C       TCMAX    - maximum decay time.
C       Tc       - counting time.
C
C
C       Start program execution.
C
        CALL REDAT
        CALL SELECT
        CALL LIMIT
        CALL STIME
        DO 1 I=1,3
 1      CC(I)=0.0
        CALL RESPON
C
C       Write the coordinates of the initial simplex.
C
        WRITE(2,5)
 5      FORMAT(' THE COORDINATES OF THE INITIAL SIMPLEX ARE: '/10X,
       +'Vertex #', 5X, 'Decay Time', 5X, 'Irrad. Time', 5X, Det. Limit'/)
        WRITE(2,6)(I,TD(I),R(I),I=1,3)
 6      FORMAT(14X,14,5X,E10.4,5X,E10.4,6X,E10.4/)
C
C       Specify the size of the standard deviation in the detection limit
```

```
C       of the vertices which requires the simplex moves to stop.
C
 8      WRITE(1,9)
 9      FORMAT(' Please specify the size of standard deviation in the '/1x,
       +'detection limit to halt the simplex moves'/)
        READ(1,12,ERR=8)HALT4
 12     FORMAT(E10.4)
C
C       Sort the simplex vertices.
C
 10     CALL SORT
        HALT2=STDEV(R)
C
C       Has the optimum been attained?
C
        IF(HALT2.LT.HALT4) GO TO 90
C
C       If the optimum has not yet been attained, reflect the simplex.
C
 20     CALL REFLEK(TDH,TRH,TDR,TRR,RR,PPR,CCR)
C
C       Compare the reflected point to the best point; if the reflected
C       point is better,then expand the simplex.
C
        PTR=RR-R(1)
        IR(PTR)30,50,50
```

```
 30     CALL EXPAND(TDR,TRR,TDH,TRH,TDE,TRE,RE,PPE,CCE)
C
C       Compare the expanded point to the best point; if it is better,
C       retain it for the new simplex & check if the optimum has been
C       attained.
C
        PTE=RE-R(1)
        IF(PTE)40,60,60
 40     R(3)=RE
        TD(3)=TDE
        TR(3)=TRE
        PPKC(3)=PPE
        CC(3)=CCE
        GO TO 10
C
C       If the reflected or the expanded point was worse than the best
C       point,compare it with the second best point and if it is better
C       than the second point,retain it for the new simplex.  Then check
C       if the optimum has been attained.
C
 50     PT1=RR-R(2)
        IF(PT1)60,70,70
 60     R(3)=RR
        TD(3)=TDR
        TR(3)=TRR
        PPKC(3)=PPR
```

```
      CC(3)=CCR
      GO TO 10
C
C     If the reflected point is worse than the second best point, then
C     contract the simplex.
C
 70   CALL CONTR(TDR,TRR,RR,TDH,TRH)
C
C     Check to see if the optimum has been attained.
C
      GO TO 10
C
C     Print the coordinates of the last Simplex.
C
 90   WRITE(2,100)
 100  FORMAT('THE COORDINATES OF LAST SIMPLEX ARE:'/10X,' Vertex #
     +',5X,'Decay Time',5X,'Irrad. Time',5X,'Det. Limit'/)
      WRITE(2,110)(I,TD(I),TR(I),R(I),I=3)
 110  FORMAT(14X,I4,5X,E10.4,5X,E10.4,6X,E10.4/)
      STOP
      END


      SUBROUTINE REDAT
      BYTE IFN(14)
      COMMON /B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
```

```
     +CMPE(300),ZZAAPM(300)/B4/L,II,JJ
C
C       Read the saturation activity data file.
C
        WRITE(1,10)
 10     FORMAT('ENTER FILENAME (CP/M STANDARD) :')
        READ(1,20)IFN
 20     FORMAT(40A1)
        CALL OPEN (6,IFN)
        I=1
 30     READ(6,32,END=34,ERR=36)EN(I),ACTSAM(I),HTIME(I),ZZAAPM(I)
 32     FORMAT(4E14.7)
        GO TO 38
 36     WRITE(1,33)
 33     FORMAT(' BAD DATA, IGNORED'/)
        GO TO 30
 38     I=I+1
        GO TO 30
 34     I=I-1
        L=I
        ENDFILE 6
C
C       Sort the data in order of decreasing gamma energy for an easy
C       build up of the Compton background (See SUBROUTINE SPEC).
C
        NN=L-1
```

```
      DO 40 I=1,NN
      J=I+1
      DO 50 M=J,L
      IF(EN(I).GT.EN(M)) GO TO 50
      S=EN(I)
      EN(I)=EN(M)
      EN(M)=S
      S=ACTSAM(I)
      ACTSAM(I)=ACTSAM(M)
      ACTSAM(M)=S
      S=ZZAAPM(I)
      ZZAAPM(I)=ZZAAPM(M)
      ZZAAPM(M)=S
      S=HTIME(I)
      HTIME(I)=HTIME(M)
      HTIME(M)=S
 50   CONTINUE
 40   CONTINUE
      RETURN
      END


      SUBROUTINE SELECT
      COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
     +CMPE(300),ZZAAPM(300)/B4/L,II,JJ
C
C     Select the element to optimize for.
```

```
 5      WRITE(1,10)
 10     FORMAT('ENTER THE ATOMIC # OF THE ELEMENT TO '/1X,'OPTIMIZE FOR'/)
        READ(1,20)IZ
 20     FORMAT(I7)
C
C       Choose physical unit on which to display gamma energies &
C       associated parameters so that the peak for optimization can be chosen
C       easily.  If more than 10 energies are expected it is
C       better to use the printer.
C
 30     WRITE(1,40)
 40     FORMAT(' Please specify where you want the gamma'/1x,' peaks to
       +be shown for your selection'/1x,'1   MONITOR'/1X,'2     PRINTER
       +'/)
        READ(1,20,ERR=30)JX
        IF(JX.NE.1).AND.(JX.NE.2)) GO TO 30
        J=0
        GO TO (35,45),JX
 45     WRITE(2,50)
        GO TO 55
 35     WRITE(1,50)
 50     FORMAT(1X,'ENTRY#',3X,'PRD. CODE',5X, 'SAT. ACT.',5X,'ENERGY'
       +/)
 55     DO 70 I=1,L
        IA=INT(ZZAAPM(I)/1.E4)
        IF(IA.NE.IZ) GO TO 70
```

```
      J=J+1
      GO TO (56,57),JX
56    WRITE(1,60)I,ZZAAPM(I),ACTSAM(I),EN(I)
      TO TO 70
57    WRITE(2,60)I,ZZAAPM(I),ACTSAM(I),EN(I)
60    FORMAT(1X,I3,5X,F10.0,2(5X,E12.5))
70    CONTINUE
75    IF(J.EQ.)) GO TO 90
77    WRITE(1,80)
80    FORMAT(1X,' SELECT A GAMMA ENERGY BY SPECIFYING ITS ENTRY'/1X,
     +'NUMBER'/)
      READ(1,20)II
85    WRITE(1,87)
87    FORMAT(1X,'1 CORRECT ENTRY'/1X,'2 MISTAKE IN CHOICE'/)
      READ(1,20)N
      GO TO (88,77),N
      GO TO 85
88    RETURN
90    WRITE(1,100)
100   FORMAT(1X,'APPARENTLY THE ELEMENT CHOSEN IS NOT IN SAMPLE'/1X,
     +'RESET THE COMPUTER TO EXAMINE YOUR SAMPLE COMPOSITION ENTRIES'/1X
     +,'****** IF YOU JUST MADE A MISTAKE IN ATOMIC # ENTRY TYPE 1'/)
      READ(1,20)M
      IF(M.EQ.1) GO TO 5
      GO TO 90
      END
```

```
        SUBROUTINE LIMIT
        COMMON/B3/TDMIN,TDMAX,TRMIN,TRMAX,TC,SMP
C
C       The subroutine enters the practical limitations for activity
C       calculations.
C
        WRITE(1,2)
 2      FORMAT(' When prompted, you will be required to enter'/1x,'
       +the minimum & maximum decay times (TDMIN &'/1X,'TDMAX), the
       +minimum and maximum irradiation times (TRMIN &'/1X,'TRMAX) as well
       +as the counting time TC'/)
        PAUSE LIMITS
 100    WRITE(1,10)
 10     FORMAT(1X,'ENTER TDMIN,TDMAX,TRMIN,TRMAX & TC'/)
        READ(5,20)TDMIN,TDMAX,TRMIN,TRMAX,TC
 20     FORMAT(5E12.5)
 200    WRITE(1,30)
 30     FORMAT(1X,'1 CORRECT ENTRY'/1X,'2  MISTAKEN ENTRY'/)
        READ(1,40)N
 40     FORMAT(I2)
        TO TO (300,100),N
        GO TO 200
 300    WRITE(1,50)
 50     FORMAT(1X,'SPECIFY THE SAMPLE SIZE IN GRAMS'/)
        READ(1,60)SMP
 60     FORMAT(E12.5)
```

```
      WRITE(1,30)
 400  READ(1,40)N1
      GO TO (500,300),N1
      GO TO 400
 500  RETURN
      END


      SUBROUTINE STIME
      COMMON/B2/TD(3),TR(3),R(3),PPKC(3),CC(3)
C
C     The subroutine reads in the initial simplex times.
C
      WRITE(1,100)
 100  FORMAT(' When prompted please enter the initial times'/1x,' to
     +define the initial simplex'/)
      PAUSE STIMES
 10   DO 20 I=L,3
      WRITE(1,30)I
 30   FORMAT(1X,'ENTER TD & TR PAIR #', I2,' IN MINUTES'/)
 20   READ(1,40)TD(I),TR(I)
 40   FORMAT(2E12.5)
      WRITE(1,50)
 50   FORMAT(1X,'PAIR #',5X,'DECAY TIME',5X,'IRRAD. TIME'/)
 55   WRITE(1,60)(I,TD(I),TR(I),I=1,3)
 60   FORMAT(2X,I2,2(5X,E12.5))
 65   WRITE(1,70)
```

```
 70     FORMAT(1X,'1   ALL TIMES CORRECT'/1X,'2   CORRECTIONS NEEDED'/)
        READ(1,80)N
 80     FORMAT(I2)
        GO TO (90,10),N
        GO TO 65
 90     RETURN
        END


        SUBROUTINE RESPON
        COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
       +CMPE(300),ZZAAPM(300)/B2/TD(3),TR(3),R(3),PPKC(3),CC(3)/B3/TDMIN
       +,TDMAX,TRMIN,TRMAX,TC,SMP/B4/L,II,JJ
C
C       The subroutine determines the response at the initial vertices.
C
 15     WRITE(2,500)ZZAAPM(ii),II,EN(II),SMP,TC,HTIME(II)
 500    FORMAT(15X,'Optimizing for ISOTOPE CODE',F8.0//1X,'PEAK # IS ',
       +I3,' & HAS A GAMMA ENERGY OF ',F7.2,'KEV.'///1X,'THE SAMPLE SIZE
       +WAS',E12.5,'GRAMS'/10X,' THE COUNTING TIME WAS', F7.2,' MINUTES'
       +/10X,' & THE ISOTOPE HALFLIFE IS',E12.5,' MINUTES'/)
        DO 50 I=1,3
 24     CALL PEAKC(TD(I),TR(I))
        WRITE(1,23)I,PKC(II)
 23     FORMAT(' VERTEX #', I3,' PEAK COUNT IS', E12.5/)
        IF(PCK(II).GT.1.) TO TO 25
        WRITE(1,26)TD(I),TR(I)
```

```
 26    FORMAT('THE SET OF TIMES TD & TR GIVE NEAR ZERO'/1X,'PEAK COUNTS
      +PLEASE TRY ANOTHER TD & TR'/1X,' CURRENT VALUES ARE TD =',E12.5,
      +'TR =',E12.5/)
       READ(1,27)TD(I),TR(I)
 27    FORMAT(2E12.5)
       TO TO 24
 25    PPKC(I)=PKC(II)
C
C      Determine FWHM
C
 40    EG=EN(II)*1.E-3
       FWHM=.844*SQRT(1.885+1.85*EG+0.907*EG**2)
C
C      Peak assumed to span 6*FWHM; the total Compton is the product of this #
C      and the Compton counts per Kev. under the peak.
C
       CBK=3.*FWHM*CMP(JJ)
C
C      Define a function proportional to detection limit.
C
       R(I)=(2.71+4.65*SQRT(CBK))/PKC(II)
       CC(I)=CBK
 50    CONTINUE
       RETURN
       END
```

```
      SUBROUTINE SORT
      COMMON/B2/TD(3),TR(3),R(3),PPKC(3),CC(3)/B4/L,II,JJ
C
C     The subroutine sorts the vertices of the simplex into best,
C     second best and worst vertex.
C
      DO 20 I=1,2
      J=I+1
      DO 10 M=J,3
      IF(R(I).LT.R(M)) GO TO 10
      S=R(I)
      R(I)=R(M)
      R(M)=S
      S=TD(I)
      TD(I)=TD(M)
      TD(M)=S
      S=TR(I)
      TR(I)=TR(M)
      TR(M)=S
      S=PPKC(I)
      PPCK(I)=PPKC(M)
      PPKC(M)=S
      S=CC(I)
      CC(I)=CC(M)
      CC(M)=S
 10   CONTINUE
```

```
 20     CONTINUE
        RETURN
        END


        SUBROUTINE SPEC
        COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
       +CMPE(300),ZZAAPM(300)/B3/TDMIN,TDMAX,TRMIN,TRMAX,TC,SMP/B4/L,
       +II,JJ
C
C       The subroutine finds the Compton edge, Compton count per Kev &
C       the photopeak count.
C
        DO 50 I=1,L
        EG=EN(I)*1.E-3
C
C       Find the photofraction efficiency of the detector.  N.B. EN(I)
C       above was multiplied by 1.E-3 because the calibration was performed
C       in Mev. whereas EN(I) is in Kev.
C
        A0=ALOG(EG)
        A1=EXP(-2.10)
        A2=EXP(-.854*A0)
        A3=EXP(-.043*A0**2)
        PF=A1*A2*A3
C
C       Find the Compton edge for gamma ray.
```

```
C
 40     CMPE(I)=EN(I)/(1.+255.5/EN(I))
        J1=I-1
C
C       Find the Compton background.
C
        IF(I.EQ.1) CMP(I)=PKC(I)*(1.-PF)/CMPE(I)
        IF(I.NE.1) CMP(I)=PKC(I)*(1.-PF)/CMPE(I)+CMP(J1)
C
C       Determine actual photocounts.
C
        PKC(I)=PKC(I)*PF
 50     CONTINUE
        DO 100 I=1,L
        IF(EN(II).LT.CMPE(I)) GO TO 100
        GO TO 110
 100    CONTINUE
 110    JJ=I-1
        RETURN
        END


        SUBROUTINE PEAKC(TDE,TIR)
        COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
       +CMPE(300),ZZAAPM(300)/B3/TDMIN,TDMAX,TRMIN,TRMAX,TC,SMP/B4/L,
       +II,JJ
C
```

```
C       The program calculates the peak count for each gamma ray
C       entering the detector.
C
 45     DO 80 I=1.L
        EG=EN(I)*1.E-3
C
C       Determine the total detector efficiency (in %) of the gamma rays.
C
        DE=.9895-.513*EXP(-EG)+1.57*(EXP-EG))**2
C
C       Since the detector calibration was performed for a constant
C       geometry, and the total detector calculation did not implement the
C       solid angle correction, the geometrical factor (GF) is taken to
C       be 1.
C
        GF=1.0
 70     PKC(I)=ACTSAM(I)*GF*DE*.01
 80     CONTINUE
C
C       Determine the correction factors viz.; saturation factor,
C       decay factor & prediction factor.
C
        DO 120 I=1.L
        HL=HTIME(I)
        PRESAT=EXP(-ALOG(2.)*TIR/HL)
        SAT=1.-PRESAT
        DECAY=EXP(-ALOG(2.)*TDE/HL)
```

```
      PREDTC=1.-EXP(-ALOG(2.)*TC/HL)
      DTC=HL*60/ALOG(2.)*PREDTC
      TGAMMA=PKC(I)
      CRECT=SAT*DECAY
      CRECT=CRECT*DTC
      PKC(I)=TGAMMA*CRECT
 120  CONTINUE
C
C     Calculate the photopeak counts as determined by the size of the
C     sample (SMP).
C
      DO 200 I=1,L
 200  PKC(I)=PKC(I)*SMP
      CALL SPEC
      RETURN
      END


      FUNCTION STDEV(R)
      DIMENSION R(3)
      COMMON/B4/L,II,JJ
C
C     The function determines the standard deviation of the response
C     function at the vertices of every simplex.  The magnitude of the
C     standard deviation is used to decide when to halt simplex
C     translation.
C
```

```
C       Determine the mean (i.e., average of the response function.
C
        SUM=0.
        DO 10 I=1,3
 10     SUM=R(I)+SUM
        AVR=SUM/3.0
C
C       Determine the standard deviation.
C
        SVAR=0.
        DO 20 I=1,3
        VAR=(AVR-R(I))**2
 20     SVAR=SVAR+VAR
        SVAR=SVAR/2.0
        STDEV=SQRT(SVAR/3.0)
        RETURN
        END


        SUBROUTINE REFLEK(TDH,TRH,TDR,TRR,RR,PPR,CCR)
        COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
       +CMPE(300),ZZAAPM(300)/B2/TD(3),TR(3),R(3),PPKC(3),CC(3)/B3/TDMIN
       +TDMAX,TRMIN,TRMAX,TC,SMP/B4/L,II,JJ
C
C       The subroutine reflects the worst point R(3) across the hyperface
C       (TDH,TRH) & determines the variables at reflected point (TDR,
C       TRR,RR,PPR,CCR)
```

```
C
C       Find the hyperface.
C
        TDH=(TD(1)+TD(2))/2.
        TRH=(TR(1)+TR(2))/2.
C
C       Determine the reflected point times.
C
        TDR=2.0*TDH-TD(3)
        TRR=2.0*TRH-TR(3)
C
C       Apply time limitations on reflected point times.
C
        IF(TDR.LT.TDMIN) GO TO 40
        IF(TDR.GT.TDMAX) GO TO 40
        IF(TRR.LT.TRMIN) GO TO 40
        IF(TRR.GT.TRMAX) TO TO 40
C
C       Determine the rest of the reflected point variables (RR,PPR,CCR)
C
        CALL PEAKC(TDR,TRR)
        PPR=PKC(II)
 20     EG=EN(II)*1.E-3
        FWHM=.844*SQRT(1.885+1.85*EG+.907*EG**2)
        CBK=3.*FWHM*CMP(JJ)
        RR=(2.71+4.65*SQRT(CBK))/PKC(II)
```

```
      CCR=CBK
 30   RETURN
C
C     Assign a very undersirable response of a reflected outside the
C     boundary condition.
C
 40   RR=100.
      PPR=0.
      CCR=0.
      RETURN
      END


      SUBROUTINE EXPAND(TDR,TRR,TDH,TRH,TDE,TRE,RE,PPE,CCE)
      COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
     +CMPE(300),ZZAAPM(300)/B2/TD(3),TR(3),R(3),PPKC(3),CC(3)/B3/TDMIN
     +TDMAX,TRMIN,TRMAX,TC,SMP/B4/L,II,JJ
C
C     The subroutine expands the reflected point (TDR,TRR) & determines
C     the expanded point variables (TDE,TRE,RE,PPE,CCE)
C     Find the expanded point times.
C
      TDE=3.*TDR-2.*TDH
      TRE=3.*TRR-2.*TRH
C
C     Apply the time limitations.
C
```

```
      IF(TDE.LT.TDMIN) GO TO 40
      IF(TDE.GT.TDMAX) GO TO 40
      IF(TRE.LT.TRMIN) GO TO 40
      IF(TRE.GT.TRMAX) GO TO 40
C
C     Determine the parameters of the expanded point.
C
      CALL PEAKC(TDE,TRE)
      PPE=PKC(II)
 20   EG=EN(II)*1.E-3
      FWHM=.844*SQRT(1.885+1.85*EG+.907*EG**2)
      CBK=3.*FWHM*CMP(JJ)
      RE=(2.71+4.65*SQRT(CBK))/PKC(II)
      CCE=CBK
 30   RETURN
C
C     Assign a very undesirable response to an expanded point outside
C     the boundary times.
C
 40   RE=100.
      PPE=0.
      CCE=0.
      RETURN
      END
```

```
      SUBROUTINE CONTR(TDR,TRR,RR,TDH,TRH)
      COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
     +CMPE(300),ZZAAPM(300)/B2/TD(3),TR(3),R(3),PPKC(3),CC(3)/B3/TDMIN,
     +TDMAX,TRMIN,TRMAX,TC,SMP/B4/L,II,JJ
C
C     The subroutine contracts the simplex & sometimes shrinks (i.e.
C     massive contracts) it.
C     Compare the reflected point to the worst point to ascertain the
C     type of contraction required.
C
      IF(RR.LT.R(3)) GO TO 10
C
C     Contract about the worst point because a more desirable response
C
      TDC=TDH-0.5*(TDH-TD(3))
      TRC=TRH-0.5*(TRH-TR(3))
      TO TO 15
C
C     Contract about the reflected point because it has better response
C
 10   TDC=TDH+.5*(TDH-TD(3))
      TRC=TRH+.5*(TRH-TR(3))
C
C     Since we are contracting about the reflected point, we now define
C     it as the current worst point because it has been shown to have a
C     better response.
```

```
C
      R(3)=RR
      TD(3)=TDR
      TR(3)=TRR
C
C     The operation of contracting a simplex occurs within the boundary
C     times hence it is not necessary to check for the violation of the
C     boundary conditions.
C
C     Determine the contracted point parameters.
C
 15   CALL PEAKC(TDC,TRC)
      PPC=PKC(II)
 30   EG=EN(II)*1.E-3
      FWHM=.844*SQRT(1.885+1.85*EG+.907*EG**2)
      CBK=3.*FWHM*CMP(JJ)
      RC=(2.71+4.65*SQRT(CBK))/PKC(II)
      CCC=CBK
C
C     Compare the contracted point to the current worst point; i.e.,
C     the point about which we contracted.
C
 40   IF(RC.GT.R(3)) GO TO 60
      TD(3)=TDC
      TR(3)=TRC
      R(3)=RC
```

```
      PPCK(3)=PPC
      CC(3)=CCC
 50   RETURN
C
C     If the contracted point was worse than worst point, shrink the simplex.
C
 60   TDS=(TD(1)+TD(2))*.5
      TRS=(TR(1)+TR(2))*.5
      CALL PEAKC(TDS,TRS)
      PPS=PKC(II)
      EG=EN(II)*1.E-3
      FWHM=.844*SQRT(1.885+1.85*EG+.907*EG**2)
      CBK=3.*FWHM*CMP(JJ)
      RS=(2.71+4.65*SQRT(CBK))/PKC(II)
      CCS=CBK
C
C     Replace the second best point by the shrink point.
C
 90   TD(2)=TDS
      R(2)=RS
      PPKC(3)=PPS
      CC(2)=CCS
      TR(2)=TRS
      RETURN
      END
```

Appendix C3

Listing of Program GRID

```
      PROGRAM GRID
      BYTE IFN(14)
      COMMON/B1/ACTSAM(300), HTIME(300), PKC(300), EN(300), CMP(300),
     +CMPE(300), ZZAAPM(300/B2/TDMIN, TDMAX, TRMIN, TRMAX, TC, SMP/
     +B3/L, II, JJ/B4/TD(400), TR(400), R(400)
C
C     The program determines the grid points for plotting the response
C     surface
C
C     ROUTINES USED IN ORDER OF CALLING.
C
C     REDAT - Enters EN, ACTSAM, HTIME and ZZAAPM
C     SELECT - Selects the element and gamma ray for optimization
C     LIMIT  - Enters the counting time and the limiting grid decay
                 and irradiation times.
C     PEAKC  - Calculates the total detected counts per gamma ray.
C     SPEC   - Calculates the spectrum.
C
C     DESCRIPTION OF ARRAYS
C
C     EN     - Energy
C     ZZAAPM - Radionuclide or Isomeric state.
C     ACTSAM - Saturation activity per gamma per gram of sample.
C     HLIME  - Halflife
C     (The above arrays are those stored on disk by SACOMP.COM).
C     PKC    - When in SUBROUTINE PEAKC, = Total counts for gamma ray;
```

```
C              - When SUBROUTINE SPEC, = Photopeak counts.
C       CMP    - Compton counts.
C       CMPE   - Compton Edge.
C       TR     - Irradiation time.
C       TD     - Decay time.
C       R      - Detection limit.
C
C       DESCRIPTION OF OTHER PERTINENT VARIABLES.
C
C       TRMIN  - Minimum irradiation time.
C       TRMAX  - Maximum irradiation time.
C       TDMIN  - Minimum decay time.
C       TDMAX  - Maximum decay time.
C       TC     - Counting time.
C       SMP    - Sample size.
C
C       Start program execution.
C
        CALL REDAT
        CALL SELECT
        CALL LIMIT
C
C       Set the steps of decay & irradiation time.
C
        WRITE(1, 10)
 10     FORMAT(1X, '# OF STEPS FOR IRRAD. & DECAY TIME'/)
```

```
      READ(1, 20) N,N1
 20   FORMAT(2I3)
      STR = (TRMAX-TRMIN)/FLOAT(N-1)
      STD=(TDMAX-TDMIN)/FLOAT(N1-1)
C
C     Clear grid arrays.
C
      DO 25 I=1, 400
      TD(I)=0.0
      TR(I)=0.0
 25   R(I)=0.0
C
C     Initialise grid point counter.
C
      J=0
C
C     Enter detection limit calculating loop.
C
      DO 30 I=1,N
C
C     Define irradiation time value.
C
      IF(I.EQ.1)TR1=TRMIN
      IF(I.NE.1)TR1=TR1+STR
      DO 40 K=1, N1
C
```

```
C     Define decay time (TD1) and irradiation time (TR1).
C
      IF(K.EQ.1)TD1=TDMIN
      IF(K.NE.1)TD1=TD1+STD
      WRITE(1, 61)TD1, TR1
 61   FORMAT('DECAY TIME = ',E10.4,' IRRAD. TIME = ',E10.4/)
C
C     Calculate the spectrum.
C
      CALL PEAKC(TD1, TR1)
C
C     Reject points with insignificant no detected counts.
C
      IF(PCK(II).LT.1.0) GO TO 40
      J=J+1
C
C     Find the Full Width at Half Maximum & the total Compton
C     background span by the peak.
C
      EG=EN(II)*1.E-3
      FWHM=.844*SQRT(1.885+1.85*EG+.097*EG**2)
      CBK=3.*FWHM*CMP(JJ)
      TD(J)=TD1
      TR(J)=TR1
C
C     Determine the detection limit.
```

```
C
      R(J)=(2.71+4.65*SQRT(CBK))/PKC(II)
 40   CONTINUE
 30   CONTINUE
C
C     Store grid data onto a disk.
C
      WRITE(1, 70)
 70   FORMAT ('ENTER FILENAME(CP/M STANDARD):'/)
      READ(1, 100)IFN
 100  FORMAT (40A1)
      CALL OPEN(6, IFN)
      DO 110 I=1,J
 110  WRITE(6, 120)TD(I),TR(I),R(I)
 120  FORMAT(3E15.5)
      ENDFILE 6
      REWIND 6
 130  STOP
      END


      SUBROUTINE REDAT
      BYTE IFN(14)
      COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
     +CMPE(300),ZZAAPM(300)/B3/L,II,JJ
C
C     Read the saturation activity data file.
```

```
C
      WRITE(1,10)
 10   FORMAT('ENTER FILENAME (CP/M STANDARD):')
      READ(1, 20)IFN
 20   FORMAT(40A1)
      CALL OPEN(6,IFN)
      I=1
 25   READ(6, 30, END=38,ERR=32)EN(I),ACTSAM(I),HTIME(I),ZZAAPM(I)
 30   FORMAT(4E14.7)
      GO TO 34
 32   WRITE(1,33)
 33   FORMAT('BAD DATA, IGNORED'/)
      GO TO 25
 34   I=I+1
      GO TO 25
 38   I=I-1
      L=I
      ENDFILE 6
C
C     Sort data in order of decreasing gamma energy.
C
      NN=L-1
      DO 40 I=1,NN
      J=I+1
      DO 50 M=J,L
      IF(EN(I).GT.EN(M)) TO TO 50
```

```
      S=EN(I)
      EN(I)=EN(M)
      EN(M)=S
      S=ACTSAM(I)
      ACTSAM(I)=ACTSAM(M)
      ACTSAM(M)=S
      S=ZZAAPM(I)
      ZZAAPM(I)=ZZAAPM(M)
      ZZAAPM(M)=S
      S=HTIME(I)
      HTIME(I)=HTIME(M)
      HTIME(M)=S
 50   CONTINUE
 40   CONTINUE
      RETURN
      END


      SUBROUTINE SELECT
      COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),COMP(300),
      CMPE(300),ZZAAPM(300)/B3/L,II,JJ
C
C     Select element to optimize for.
C
 5    WRITE(1,10)
 10   FORMAT(' OPTIMIZATION ELEMENT ATOMIC NUMBER?'/)
      READ(1,20)IZ
```

```
20     FORMAT(I3)
       J=0
       WRITE(1,25)
25     FORMAT(1X,'ENTRY#',3X,'PRD. CODE',5X, 'SAT.ACT.', 5X, 'ENERGY'/)
       DO 30 I=1,L
       IA=INT(ZZAAPM(I)/1.E4)
       IF(IA.NE.IZ) GO TO 30
       J=J+1
       WRITE(1,40)I,ZZAAPM(I),ACTSAM(I),EN(I)
40     FORMAT(1X,13,5X,F10.0,2(5X,E12.5))
30     CONTINUE
       IF(J.EQ.0) GO TO 90
55     WRITE(1,50)
50     FORMAT(1X,'GAMMA ENERGY BY ENTRY # OF INTEREST'/)
       READ(1,20)II
65     WRITE(1,70)
70     FORMAT(1X,'1 CORRECT ENTRY'/1X,'2 MISTAKE IN CHOICE'/)
       READ(1,20)N
       GO TO (80,55),N
       GO TO 65
80     RETURN
90     WRITE(1,100)
100    FORMAT(1X, 'APPARENTLY THE ELEMENT CHOSEN IS NOT IN SAMPLE'/1X,'
      +RESET THE COMUPTER TO EXAMINE YOUR SAMPLE COMPOSITION ENTRIES'/,
      +******IF YOU JUST MADE A MISTAKE IN ATOMIC # ENTRY TYPE 1'/)
       READ(1, 20)M
```

```
      IF(M.EQ.1) TO TO 5
      GO TO 90
      END


      SUBROUTINE LIMIT
      COMMON/B2/TDMIN,TDMAX,TRMIN,TRMAX,TC,SMP/B3/L,II,JJ
C
C     The subroutine enters the practical limitations for activity
C     calculations.
C
 100  WRITE(1,10)
 10   FORMAT(1X,'ENTER TDMIN,TDMAX,TRMIN,TRMAX & TC'/)
      READ(5,20)TDMIN,TDMAX,TRMIN,TRMAX,TC
 20   FORMAT(5E12.5)
 200  WRITE(1, 30)
 30   FORMAT(1X,'1 CORRECT ENTRY'/1X,'2 MISTAKEN ENTRY'/)
      READ(1,40)N
 40   FORMAT(I2)
      IF((N.NE.1). AND.(N.NE.2)) TO TO 200
      IF(N.EQ.2) GO TO 100
 305  WRITE(1,55)
 55   FORMAT('PLEASE ENTER THE SAMPLE SIZE IN GRAMS'/)
      READ(1,60)SMP
 60   FORMAT(2E12.5)
      WRITE(1,30)
 400  READ(1,40)N1
```

```
      GO TO (500,305),N1
      TO TO 400
 500  RETURN
      END


      SUBROUTINE SPEC
      COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
     +CMPE(300),ZZAAPM(300)/B2/TDMIN,TDMAX,TRMIN,TRMAX,TC,SMP/B3/L,
     +II,JJ
C
C     The subroutine finds the Compton edge, Compton count per Kev &
C     the photopeak count.
C
      DO 50 I=1,L
C
C     Since the calibration parameters were obtained in Mev. convert
C     gamma energy units to Mev.
C
      EG=EN(I)*1.E-3
C
C     Determine photofraction of the peak.
C
      AO=ALOG(EG)
      PF=EXP(-2.1-0.854*AO-0.043**2)
C
C     Find the Compton edge for each gamma ray.
```

```
C
 40     CMPE(I)=EN(I)/(1.+255.5/EN(I))
        J1=I-1
C
C       Find the Compton background.
C
        IF(I.EQ.1) CMP(I)=PKC(I)*(1.-PF)/CMPE(I)
        IF(I.NE.1) CMP(I)=PKC(I)*(1.-PF)/CMPE(I)+CMP(J1)
C
C       Determine actual photocounts.
C
        PKC(I)=PKC(I)*PF
 50     CONTINUE
        DO 100 I=1,L
        IF(EN(II).LT.CMPE(I)) GO TO 100
        GO TO 110
 100    CONTINUE
 110    JJ=I-1
        RETURN
        END


        SUBROUTINE PEAKC(TDE,TIR)
        COMMON/B1/ACTSAM(300),HTIME(300),PKC(300),EN(300),CMP(300),
       +CMPE(300),ZZAAPM(300)/B2/TDMIN,TDMAX,TRMIN,TRMAX,TC,SMP/B3/L,
       +II,JJ
C
```

```
C       The subroutine calculates the peak count for each gamma ray
        entering the  detector.
C
C       Determine saturation activity with detector efficiency
C       correction.
 45     DO 80 I=1,L
C
C       Define the total detector efficiency.
C
        EG=EN(I)*1.E-3
        DE=.9895-.513*EXP(-EG)+1.57*(EXP(-EG))**2
C
C       The detector efficiency was measured at a constant distance
C       (lowest shelf) therefore we define the geometrical efficiency
C       as unity.
C
        GF=1.0
 70     PKC(I)=ACTSAM(I)*DE*.01
 80     CONTINUE
C
C       Determine the correction factors viz.; saturation factor,
C       decay factor & prediction factor.
C
        DO 120 I=1,L
        HL=HTIME(I)
        PRESAT=EXP(-ALOG(2.)*TIR/HL)
```

```
      SAT=1.-PRESAT
      DECAY=EXP(-ALOG(2.)*TDE/HL)
      PREDTC=1.-EXP(-ALOG(2.)*TC/HL)
      DTC=HL*60/ALOG(2.)*PREDTC
      TGAMMA=PKC(I)
      CRECT=SAT*DECAY
      CRECT=CRECT*DTC
      PCK(I)=TGAMMA*CRECT
  120 CONTINUE
C
C     Calculate the total counts for each gamma ray as determined by
C     the scaling factor (i.e. sample size SMP)
C
      DO 200 I=1,L
  200 PKC(I)=PKC(I)*SMP
      CALL SPEC
      RETURN
      END
```

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