An MS-DOS-Based Program for Analyzing Plutonium Gamma-Ray Spectra

Volume 1 Data-Analysis Methodology and Software

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it was necessary to obtain results in the field. Six years ago, before the advent of powerful personal computers, we developed a portable (weighing 24 lb and sized to fit under an airline seat), microcomputer-based unit for the IAEA that could analyze a plutonium gamma-ray spectrum for plutonium isotopes in approximately one minute. A detailed report of this unit and its operation is given in Ref. 1. When IAEA inspectors began to use portable, MS-DOS-based personal computers, we were asked to transfer the plutonium gamma-ray analysis software to these computers and to upgrade it so that it could also be used to analyze gamma-ray spectra taken of mixed uranium and plutonium oxides.

The spectral-analysis problem requires sophisticated data-analysis techniques, but considering the practical aspects of inspector use, the analysis programs must be user friendly. Only the spectrum filename and the positions of the 148-keV ^{241}Pu and 208-keV ^{237}U peaks need to be entered by the inspector to initiate a complete analysis. The analysis determines plutonium isotopic ratios from gamma-ray peaks in the 110- to 390-keV region of the spectral data. This requires proper descriptions of the peak shape and background continuum and a method for unfolding peak multiplets.

Gamma-ray peak profiles are generated for each peak group by using known gamma-ray energies and peak shapes. The peak shapes are determined from the 148- and 208-keV gamma-ray peaks in the spectrum, and this information is parameterized as a function of energy. The peak response profiles are fitted to the observed multiplets by means of a linear least-squares technique to determine the areas of the individual peaks. Therefore, all essential information is obtained once the locations of the two peak positions are specified.

In addition to the data-analysis program, data-transfer and data quality-assurance programs are provided. The user interacts with these programs by means of menus and screens displayed on the computer: menus for selection of desired action (data transfer, complete sample-description form, etc.), and screens for obtaining information required for a desired action (peak positions, filename, declared plutonium isotopic abundances, etc.). The user-interface (menus and screens) software uses a compiled version of dBASE III (i.e., Clipper), as specified in Chapter V.a of Ref. 2. The data-transfer, analysis, and quality-assurance applications are written in FORTRAN.

2. Data-Analysis Methodology

The general method for determining plutonium isotopic-abundance ratios using high-resolution gamma-ray spectrometry is based on a principle commonly expressed by the following equation:

$$\frac{N_i}{N_j} = \frac{I_i}{I_j} \cdot \frac{B_j}{B_i} \cdot \frac{e_j}{e_i} \cdot \frac{t_i}{t_j} \quad (1)$$

where N_i/N_j is the atom ratio of isotopes i and j , I is the observed peak intensity, B is the gamma-ray branching intensity, e the counting efficiency, and t the half-life. The branching intensities and half-lives are known, and the peak intensities are measured, so the only remaining unknowns are the efficiencies. The influence of detector-counting efficiency and attenuation, physical factors that are functions of energy, is minimized by taking ratios of gamma-ray peak intensities for pairs of peaks that are nearly equal in energy. Small differences can be determined accurately by using a curve established from other neighboring peaks that are caused by gamma emissions of known relative intensities; this technique is commonly known as the intrinsic calibration method.³

Determination of peak intensities must be precise to give accurate final results. A careful analysis and interpretation of gamma-ray peaks includes the proper delineation of the peak shape, a reproducible description of the background continuum under the peak or peak group, and a method for

unfolding peak multiplets and removing small interferences. The basic equation for describing peak shapes, developed by Gunnink,⁴ is written as follows:

$$y_i = y_0 e^{\alpha(x_i - x_0)^2} + A y_0 e^{B(x_i - x_0)} [1 - e^{C\alpha(x_i - x_0)^2}]^\delta, \quad (2)$$

where

y_i = net data counts

y_0 = peak height

α = peak-width parameter

x_i = channel value of the i th point

x_0 = position of the peak's centroid

A, B, C = shape parameters that describe the tailing function

$\delta = 1$ for $x_i - x_0 < 0$

$\delta = 0$ for $x_i - x_0 > 0$.

Six parameters— y_0 , x_0 , α , A , B , and C —characterize a particular gamma-ray peak (see Fig. 1). The parameters α , A , B , and C are needed to describe the peak shape but normally are not of great interest by themselves. The peak-fitting process is simplified if their values are predetermined from the given spectrum and then treated as fixed, rather than free parameters.

Figure 1. The mathematical components used to describe the shape of a gamma-ray peak.

The peak width is commonly specified by the full width at half maximum (FWHM = 2.355σ). This is related to α by

$$\alpha = 1.0/2\sigma^2 \quad (3)$$

The width of a peak results primarily from summing the instrumental noise (Γ_n) and an energy-dependent term (Γ_s) resulting from statistical fluctuations in the number of ion pairs formed in the detector. To a first approximation, the statistical term is linear with energy, and the noise contribution is constant. Thus, the observed total peak width (Γ_t) may be expressed by

$$\Gamma_t^2 = \Gamma_n^2 + \Gamma_s^2 = K_1 + K_2 E \quad (4)$$

FWHM is related to energy by

$$[(FWHM)^2 - 0.462] \cdot (GAIN)^2 = K_1 + K_2 E \quad (5)$$

The values of K_1 and K_2 can be calculated by determining the FWHM of low- and high-energy peaks in a spectrum. The value 0.462 is a Sheppard's correction for the error that results from treating all the counts in a given channel as if they were concentrated at the channel center.

The parameters A and B characterize the amplitude and slope, respectively, of an exponentially rising term that describes the tail that occurs on the low-energy side of a gamma-ray peak. This term is multiplied by $1 - \exp[C \alpha(x - x_0)^2]$ to reduce its contribution to zero at the peak centroid. The quantity C is a rather insensitive parameter, and a value of 0.4 appears suitable for all germanium-detector systems.

The tail-slope parameter, B , is detector-dependent only, whereas the tail-amplitude parameter, A , is both detector- and energy-dependent. A simple algorithm,

$$\ln A = C_1 + C_2 E \quad (6)$$

describes the approximate relationship between A and the gamma-ray energy. As in the case for α , two or more peaks can be used to evaluate the constants C_1 and C_2 in this expression. Therefore, the peak shapes for a given spectrum can now be predetermined from two peaks and specified in terms of five parameters: K_1 and K_2 (used to determine α), C_1 and C_2 (used to calculate A), and B .

To determine these parameters, the 148.6-keV peak of ^{241}Pu and the 208-keV peak of ^{237}U are used. Usually, these two peaks are the most intense peaks in the spectrum, especially for high-burnup plutonium. Good statistical significance is required to accurately determine A 's energy dependence, and in most cases, the spectra encountered are not statistically good. In addition, the 148.6-keV peak also has interferences on the low-energy side, which increase the uncertainty in A at that energy. Because changes in A over an interval of 100 keV are 1% or less, the analysis code determines A and B from the 208-keV peak and treats A as a constant for the spectrum. The area of the tail is not included in the total area of the peak in this analysis.

The 148.6- and 208-keV peaks are also used as references for determining the gain and channel-zero energy of the spectrum. Using this information (assuming the conversion gain is linear), the exact positions of the peaks to be analyzed can be computed. If the value of x_0 in Eq. (2) is also known (in addition to the other shape parameters), the only free parameter remaining is the peak height. The resulting equation becomes linear in form and can be solved by a linear least-squares method rather than by an iterative technique.

Data points, Y_i , in an overlapping peak multiplet are considered linear combinations of the appropriate contribution from each peak, j , so that

$$Y_i = \sum_{j=1}^n y_{0j} \cdot f(j, \text{peak parameters}) \quad (7)$$

However, not all of the peak heights in a group need to be free parameters because peaks belonging to a given isotope can be related by their known branching intensities. This allows for removal of interferences in cases where peaks overlap severely.

Before determining the net area of a peak, the background beneath the peak or peak group must be removed. This is usually done by interpolating the background or by using a straight line or a polynomial function. However, such a procedure is, in principle, incorrect and may lead to erroneous results. Thus, the following explicit function is used to interpolate background values beneath the peak group:

$$B_i = b_n + (b_m - b_n) \cdot \left(\sum_{j=n}^{j=i} Y_j \right) / \left(\sum_{k=n}^{k=m} Y_k \right) \quad (8)$$

where

B_i = computed background at channel i

Y_i = spectrum count of channel i

b_n = average background level of the low-energy side of the peak

b_m = average background level of the high-energy side of the peak.

This function produces smoothed background steps that occur at each peak position. It works equally well for single peaks and complex peak groups.

Energy Regions

The choice of gamma-ray peaks for analysis to determine the desired isotopic ratios was limited to the energy ranges of the spectra normally taken by the IAEA inspectors. The energy ranges usually measured are from 110 to 220 keV or from 110 to 400 keV. In the energy interval from 110 to 220 keV, there are nine energy regions containing one or more peaks that are analyzed by the data-analysis code. The energies and intensities of the gamma rays analyzed in these regions are listed in Table 1. Most of the energy and intensity information used in the data-analysis program is taken from a report published by Gunnink.⁵ In certain instances, however, changes have been made to reflect results obtained by others^{6,7} and ourselves. NOTE that this analysis assumes that ^{237}U is in secular equilibrium with its parent, ^{241}Pu . The details of analyzing these regions follow.

Region 1. 122 to 130 keV

Gamma rays from ^{239}Pu and ^{241}Am exist in this region. The 125.29- and 129.29-keV peaks contribute to the determination of the $^{241}\text{Am}/^{239}\text{Pu}$ ratio. The 125.29-keV peak is a member of an unresolved triplet whose other components are the ^{239}Pu peaks at 124.51 and 125.21 keV. These interferences in the ^{241}Am 125.29-keV peak are removed by ratioing the ^{239}Pu 124.51- and 125.21-keV peaks to the ^{239}Pu 129.29-keV peak. The ^{239}Pu 123.62-keV peak is also ratioed to the 129.29-keV peak. The ratios are determined from the branching intensities of these peaks.

These ratios are incorrect to the degree that the detection efficiency differs between the lower-energy peaks and the 129.29-keV peak. Because the relative detection efficiencies cannot be

established until all of the peak areas are determined, it is impossible to correct these ratios at this point in the analysis. Corrections to these ratios are made later, when the isotopic abundances are determined.

A difficulty in the analysis of this region is how to determine the background below (on the low-energy side of) this region. This is caused by the close proximity of the plutonium $K\beta$ x rays. The program searches for the channel with the minimum counts between the 123.6- and 124.5-keV peaks and uses the minimum count value as the low-energy background level for this region.

Region 2. 141 to 151 keV

This region contains gamma rays from ^{239}Pu , ^{241}Pu , and ^{241}Am . Although there are eight peaks in this region, the only information used from this region is that of the ^{241}Pu 148.6-keV peak area. The other peaks are included in the analysis only to determine a satisfactory background for the 148-keV peak. The weak ^{239}Pu 146.1-keV peak is ratioed to the 144.21-keV peak. The weak ^{241}Am 150.1-keV peak is ratioed to the ^{241}Am 146.6-keV peak. Again, any corrections to this ratio for relative detection-efficiency differences are made later.

Region 3. 152 to 154 keV

A gamma ray from ^{238}Pu exists in this region. The intensity listed in Table 1 is 3.2% smaller than the value given by Gunnink.⁵ In addition, a sum peak can occur at 154.2 keV because of the summing of the ^{241}Am 59.54-keV gamma ray with the uranium 94.66-keV $K\alpha$ x ray. The existence of this sum peak may compromise the determination of the high-energy background for the 152.7-keV peak. Every effort should be made to eliminate this sum peak and others (see next region) with the proper combination of absorbers.

Region 4. 159 to 166 keV

Gamma rays from ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{237}U , and ^{241}Am are analyzed in this region. Note that the energy of the ^{240}Pu gamma ray is different from that given by Gunnink.⁵

A single peak from this region is used to determine the ^{240}Pu abundance. The 160.313-keV gamma ray from ^{240}Pu forms an unresolved multiplet with the 159.96- and 160.00-keV peaks from ^{241}Pu and ^{239}Pu . In the analysis of this multiplet, the 160.00-keV peak is ratioed to the 161.479-keV peak.

In addition to the severe overlap of peaks in this region, the analysis is made more difficult and less accurate when sum peaks occur in this region. The sum peaks are the result of summing the ^{241}Am 59.53-keV gamma ray and the neptunium and uranium $K\beta$ x rays. One of these sum peaks directly interferes with the 160.3-keV ^{240}Pu peak, while two other sum peaks interfere with determining the low-energy background level. The possible interference with the ^{240}Pu 160.3-keV peak *cannot* be avoided or corrected for by means of data manipulation or calculation; consequently, the ^{240}Pu result will be biased if the sum peaks are not eliminated. The intensity of the ^{241}Am 59.5-keV peak should be reduced with cadmium absorbers so that the height of this peak in the spectrum is approximately equal to the height of the strongest peak in the 100-keV region.

Region 5. 184 to 187 keV

This region contains the strongest gamma ray from ^{235}U . The intensity of this gamma ray is used to determine the $^{235}\text{U}/^{239}\text{Pu}$ ratio.

Region 6. 203 to 208 keV

Gamma-ray peaks from ^{239}Pu , ^{241}Pu , ^{237}U , and ^{241}Am exist in this region as well as two peaks from ^{235}U , if present in the sample. This region is important for determining the $^{241}\text{Pu}/^{239}\text{Pu}$ ratio. For high-burnup materials, an accurate determination of this ratio from this region is difficult because the ^{239}Pu 203.6-keV peak intensity is weak, and the background beneath it is difficult to establish because of counts from the low-energy tail of the strong ^{241}Pu - ^{237}U 208-keV peak.

If the ^{235}U 185-keV gamma ray is not in the spectrum, the 203- and 208-keV peaks are integrated separately. Before integration of the 203-keV peak, the low-energy tail from the 208-keV peak is removed from beneath the 203-keV peak. If the ^{235}U 185-keV gamma ray is found in the spectrum, the 203-keV, 208-keV, and ^{235}U peak areas are determined by simply integrating the net counts in the region and proportioning the total net counts according to each individual peak height. The peak heights are determined by fitting the tops of the peaks. Again, the net counts in the 203-keV region are corrected for counts from the low-energy tail of the 208-keV peak.

The 208-keV gamma ray from ^{241}Pu - ^{237}U and ^{241}Am is normally the strongest in the spectrum. If pulse pileup resulting from high counting rates occurs, the spectral effects are readily evident on the high-energy side of the 208-keV peak. This region is examined by the program to determine if the effects of pileup are present, and reports their presence if they are found. The spectral effects of pulse pileup cause distorted peak shapes and may cause biases in determining the high-energy background level, thus leading to inaccuracies in determining the areas of the peaks. Normally, the high-energy background level is determined from a region immediately above the gamma-ray peak. In the analysis of the 208-keV peak, the high-energy background region has been moved higher in energy to avoid counts caused by pulse pileup. If the effects of pileup are present, the counting rate should be reduced, and the pole-zero adjustment should be checked. If the counting rate cannot be reduced, then a shorter amplifier time constant should be considered. However, this normally results in poorer resolution.

Region 7. 267 keV

A gamma-ray peak from ^{241}Pu - ^{237}U and ^{241}Am is present in this region. This peak is used primarily to define the shape of the relative gamma-ray detection-efficiency curve for a measurement. It is especially important if the 330- and 370-keV regions are used as described in the following paragraphs.

Two additional energy regions—332 to 345 keV and 367 to 385 keV—are included in the analysis if the gain and channel-zero energy are set to include these regions in the spectrum. Inclusion of these regions improves the precision in determining the $^{241}\text{Pu}/^{239}\text{Pu}$ and the $^{241}\text{Am}/^{239}\text{Pu}$ isotopic ratios, provided sufficient statistical precision is obtained for peaks in these regions.

Before analyzing these regions, the data-analysis program redetermines the gain and zero of the spectrum by determining the position of the ^{239}Pu 375.04-keV peak. This is done to ensure, should nonlinearity exist in the spectrum, that these energy regions are properly located by the data-analysis program. The details of analyzing these regions follow.

Region 8. 332 to 347 keV

Gamma rays from the decay of ^{239}Pu , ^{241}Pu - ^{237}U , and ^{241}Am contribute to this region. The gamma-ray peaks at 332.3 keV resulting from ^{241}Pu - ^{237}U and ^{241}Am have an interference from ^{239}Pu , which is removed in the fitting process by ratioing its intensity to the ^{239}Pu 345.0-keV peak. This ratio is corrected for efficiency differences later in the analysis.

Region 9. 336 to 385 keV

This region contains gamma rays from ^{239}Pu , ^{241}Pu - ^{237}U , and ^{241}Am . The two weaker ^{239}Pu peaks at 367 and 368.5 keV that interfere with the peak from ^{241}Pu - ^{237}U and ^{241}Am at 368.6 keV are removed by ratioing their intensities to the ^{239}Pu 375.0-keV peak.

Analysis

The data-analysis program analyzes each region separately. First, the background distribution is removed from the peak region, and then the position of a reference peak in each region is determined by fitting the top of that peak. The peaks used as reference peaks in each region are denoted by

asterisks in Table 1. The positions of the remaining peaks in each region are located relative to the reference peak by using the gain and channel-zero energy of the spectrum. With this position information and the other peak shape parameters, a response function for each peak is generated. A set of linear equations, with the peak heights as the unknowns, are solved by a simple matrix-inversion procedure. Before the least-squares fit is performed, the equations are appropriately weighted. This is done by dividing each channel-count equation by the square root of the gross channel count.

The peak areas are calculated by using the integral of a Gaussian from the peak heights and the appropriate FWHM values, which are calculated from the predetermined peak-shape parameters by using Eq. (5). The errors on the peak areas are determined from the diagonal elements of the inverted matrices. Some of the off-diagonal (covariant) elements from the inverted matrices are preserved for determining errors on some of the isotopic ratios. The errors are multiplied by the square root of the chi-square value for the fit. After analysis of each region, the results—peak energies, areas, and errors—are printed or displayed if peak areas are requested in the report. An example of the printout is shown in Fig. 2.

The next step in the data analysis is to determine isotopic ratios from the peak areas. As shown in Eq. (1), this requires corrections for the influence of detector-counting efficiency and gamma-ray attenuation (caused by the sample matrix or by external absorbing materials) on gamma-ray peak areas. Because ratios of peak areas are involved, it is not necessary to determine the corrections to the peak areas absolutely, but only the relative relationship between the corrections, which is then applied to the peak-area ratio. To minimize these corrections and, consequently, to make them less sensitive to errors in their determination, the peak-area ratios are taken of pairs of gamma rays that are as close as possible in energy. If the energies are close, attenuation and efficiency differences are small, thus making the corrections small.

```

----- IAEA Plutonium Analysis Diagnostics -----

GROUP # 1
REDUCED CHI-SQUARE =          .8
PEAK EN.    COUNTS    %ER
122.99      1234.      6.26
123.62       17.      FIXD
124.51       54.      FIXD
125.21       49.      FIXD
125.30      6254.      1.72
129.29      5452.      1.89

GROUP # 2
REDUCED CHI-SQUARE =          1.3
PEAK EN.    COUNTS    %ER
141.57       44.    100.00
143.35       27.    100.00
143.77      240.    52.18
144.21      321.    30.18
146.08      135.      FIXD
146.56     1452.     6.58
148.57     46273.     .57
150.11      235.      FIXD

```

Figure 2. A portion of the data-analysis diagnostic printout, showing peak energies, areas, and errors.

An intrinsic calibration technique is used to determine these corrections from peak areas measured in each spectrum. Peak areas from ^{239}Pu and ^{241}Pu - ^{237}U gamma rays (denoted by * in Table 1) are corrected for gamma-ray branching intensities and normalized to the value obtained for the 208-keV peak (i.e., the relative detection efficiency value for the 208.0-keV peak is set equal to 1.0). These values are fit to the following functional form for relative efficiency:

$$\epsilon^j = \frac{\ln\left(\frac{I_i}{B_i}\right)}{\ln\left(\frac{I_{208}}{B_{208}}\right)} = a_0 + a_1 \delta_i + \sum_{j=1}^{2\text{ or }3} \{a_{j+1} [\ln E_i - \ln (0.208)]^j\} \quad (9)$$

The term E_i is the energy of the corresponding gamma ray, in MeV. The coefficient a_1 normalizes the ^{239}Pu data points to the ^{241}Pu - ^{237}U data points. If the 330- and 370-keV regions are included in the spectrum, a cubic function is fit to the data (the sum from $j = 1$ to $j = 3$). The ^{241}Pu 148.6-keV peak area is not used in determining the relative efficiency curve because it would require correction of the ^{241}Pu - ^{237}U 165-, 208-, and 267-keV peaks for the ^{241}Am contribution, which is unknown at this point in the analysis.

From this functional relationship, the necessary corrections to the peak-area ratios can be calculated. The methodology⁸ used to calculate isotopic ratios from peak areas is best illustrated by an example. Consider the 203/208-keV peak pair, which belong to ^{239}Pu , ^{241}Pu - ^{237}U , and ^{241}Am . If A^{203} and A^{208} represent the net areas of the two peaks, the following equations can be written to describe their observed values:

$$A^{203} = \epsilon^{203} B_2^{203} X_2 \quad (10)$$

and

$$A^{208} = \epsilon^{208} B_4^{208} X_4 + \epsilon^{208} B_5^{208} X_5 \quad ,$$

where components 2, 4, and 5 represent ^{239}Pu , ^{241}Pu , and ^{241}Am , respectively, and X equals the amount of each isotopic component. A single linear equation is obtained by dividing one equation by the other, which can be rewritten in the following form:

$$B_2^{203} - \frac{A^{203} \epsilon^{208} B_4^{208}}{A^{208} \epsilon^{203}} \frac{X_4}{X_2} - \frac{A^{203} \epsilon^{208}}{A^{208} \epsilon^{203}} B_5^{208} \frac{X_5}{X_2} = 0 \quad (11)$$

The ratio $\epsilon^{208}/\epsilon^{203}$ is calculated from the relative-efficiency curve given by Eq. (9). The unknowns are the ratios X_4/X_2 and X_5/X_2 , which represent disintegration ratios for $^{241}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Am}/^{239}\text{Pu}$, respectively. Equations similar to Eq. (11) are developed for the 125/129-, 148/129-, 152/148-, 160/165-, 332/345-, and 368/375-keV peak pairs. After the complete set of linear equations has been formed and weighted, it is solved by the method of least squares. The weighting of each equation involves the peak-area variances, any possible covariances between the peaks, and the relative efficiency for the energy region represented by the equation. A simple matrix-inversion procedure yields the disintegration ratios and their attendant errors.

The disintegration ratios must be corrected for isotopic half-lives and mass differences to obtain isotopic weight ratios. The half-lives used for the plutonium isotopes and ^{241}Am are listed in Table 2. The isotopic weight ratios and their absolute total errors are printed or displayed. The results also include the age of the sample since separation from ^{241}Am . This age is calculated on the basis of the

$^{241}\text{Am}/^{241}\text{Pu}$ ratio by using the standard parent-daughter relationship. This portion of the results readout is shown in Fig. 3.

The weight-percent abundance of each plutonium isotope and the ^{241}Am abundance vs total plutonium are also displayed or printed. Calculation of these abundances requires information on the ^{242}Pu abundance. Because the ^{242}Pu gamma rays are not measurable, it is necessary either to input this information, if known, or to calculate it by using a correlation. The data-analysis program has been written to allow the user to enter the ^{242}Pu weight-percent value along with its relative error, when known. If the user enters zero for the ^{242}Pu weight-percent abundance in the plutonium abundances form (see Vol. 2 for descriptions of forms), then the analysis program calculates the ^{242}Pu abundance based on a correlation⁹ that includes the ^{241}Pu abundance; i.e.,

$$[242] = \frac{K [240] [241]}{[239]^2}, \quad (12)$$

IAEAPU ANALYSIS OF SPECTRUM:sdata.bin

```

Pu238/Pu239 = .004713 +- .000030
Pu240/Pu239 = .209061 +- .001604
Pu241/Pu239 = .004713 +- .000213
Am241/Pu241 = .944704 +- .004144
U235/Pu239 = .000000 +- .000000

```

```

PLUTONIUM AGE = 5072.46 +- 16.37 DAYS
                = 13.89 +- .04 YEARS

```

	MEAS. WT.PCT.	ABS. ERR	DECAY CORR. OPERATOR WT.PCT.	(M-O) %
PU238 =	.3687 +-	.0022	.3650	1.0000
PU239 =	78.2332 +-	.1408	77.9210	.4000
PU240 =	16.3555 +-	.1055	16.6830	-1.9608
PU241 =	3.8886 +-	.0143	3.8500	1.0000
PU242 =	1.1540 +-	.1154	1.1540	.0000
AM241 =	3.6736 +-	.0068	3.5666	3.0000

```

U235/PU RATIO = .0000 +- .0000

```

```

PU240 EFFECTIVE = 19.0855 +- .2185

```

```

SPECIFIC POWER (mW/g) = 9.0888 +- .0123

```

```

VALUE OF T**2 = 27.699

```

Figure 3. A portion of the data-analysis readout, showing the final results of the analysis.

where [242], [240], [241], and [239] are the corresponding isotopic abundances. A value of 52 for K has been obtained by fitting the correlation to a set of plutonium isotopic data that was decay-corrected to the time of reactor discharge. This correlation is linear and, in general, is independent of the reactor type. Uncertainties associated with ^{242}Pu determinations obtained by using this correlation are usually less than 10% for a wide range of materials, provided the cooling time (time since reactor discharge) is known.

The ^{241}Pu abundance in Eq. (12) must be corrected for decay since removal from the reactor. The data-analysis program corrects only for decay since the last separation because additional decay as a result of cooling or other americium separations is usually unknown. Each year of cooling time for which a correction is not made results in a bias of about 5% in the ^{241}Pu abundance. If the cooling time is not known, then a default value of 4 years is used.

The specific power and "Pu-240 effective" values for the sample are also calculated and printed or displayed along with their corresponding errors. The specific power values used for each isotope are shown in Table 3.

The "Pu-240 effective" value is calculated by using the following equation:

$$\text{Pu-240 effective} = 2.49 [238] + [240] + 1.57 [242] \quad (13)$$

where [238], [240], and [242] are the corresponding isotopic abundances.

Table 2. Plutonium and americium half-lives used.

Isotope	Half-life (y)
^{239}Pu	87.74
^{240}Pu	24,119
^{241}Pu	6,542
^{241}Am	14.40
^{241}Am	433.0

Table 3. Plutonium and americium specific power values.

Isotope	Specific power (mW/g)
^{239}Pu	567.16
^{240}Pu	1.929
^{241}Pu	7.098
^{241}Pu	3.339
^{241}Am	114.23
^{242}Pu	0.1146

3. Software Descriptions

The following are brief descriptions of the software developed for the MS-DOS-based systems to analyze plutonium gamma-ray spectra for plutonium isotopic abundances. Descriptions of the data-analysis program and the various subroutines are given. Listings are provided in Vol. 2.

The software was developed for IBM-compatible computers that use MS DOS. The data-transfer, analysis, and quality-assurance applications are written in Microsoft FORTRAN-77. The menu-and-forms (see Vol. 2 for descriptions of the menus and forms) front-end program is written in Clipper-compiled dBASE III language with on-line help implemented by using Flashup.

Data-Analysis Program—IAEAPY

This program analyzes plutonium gamma-ray spectra for isotopic ratios and weight-percent abundances of total plutonium. Plutonium and ^{241}Am gamma-ray peaks from 123 to 383 keV are analyzed. It can analyze spectra contained in as many as 4096 channels. For a complete analysis, the spectrum must start at or below 121.5 keV and include the ^{241}Pu - ^{237}U 208.0-keV peak. The only input

required to the program is the spectrum filename and the positions of the ^{241}Pu 148.6-keV and ^{241}Pu - ^{237}U 208.0-keV peaks. The necessary input information is entered by means of the plutonium-analysis form (see Vol. 2). Input of the ^{242}Pu weight-percent abundance is allowed in the plutonium isotopic-abundance form, but it is not required. The remaining information necessary for analyzing the plutonium spectra is contained in the program.

The first step in the data analysis is to determine the gain (GAIN), channel-zero energy (ZERO), and the gamma-ray peak-shape parameters: SHAPC(1), SHAPC(2), ATAIL, and BTAIL. [SHAPC(1) and SHAPC(2) are the K_1 and K_2 parameters in Eq. (5); ATAIL is the tail-amplitude parameter, A , in Eq. (2); and BTAIL is the tail-slope parameter, B , in Eq. (2) times the gain.] Values for these parameters are obtained by analyzing the 148- and 208-keV reference peaks. Initially, approximate values for several parameters are used in analyzing the reference peaks, so the analysis is repeated, using the results obtained in the first pass for these parameters.

Approximate gain and zero values are determined from the user-provided reference peak locations. Before the reference peaks are analyzed for channel positions and shape parameters, the background distributions beneath the peaks are removed. The average counts (determined with the AVE subroutine) found on either side of the peak are used as the corresponding background levels. The background is subtracted from beneath the peaks by using the BKGRD subroutine.

The peak position, PKPS, and the peak-width parameter, ALFA, are determined with the GFIT2 subroutine, which fits a parabola to the natural logarithm of the net counts in the upper half of a peak. In addition to the background-corrected data in the Y array, inputs to this subroutine are the approximate position, within the Y array, of the reference peak, JPK, and the ALFA, ATAIL, and BTAIL parameters. ALFA is equal to $-4 \ln 2 / \text{FWHM}^2$, where the FWHM is expressed in channels. GFIT2 returns the peak position, the peak height PHT, and a redetermined value for the peak-width parameter ALFA. With this information, the peak-tail parameters, ATAIL and BTAIL, are determined with the CTAIL subroutine.

From the more-accurate positions of the reference peaks, new gain and zero values for the spectrum can be calculated. The tail-parameter values are checked to ensure that they fall within some reasonable bounds. Unknown peak interferences or poor counting statistics in the reference peaks could cause these values to fall out of bounds. The bounds have been empirically determined and require that the value given by BTAIL/GAIN be greater than 1.5 and less than 3.5. If BTAIL/GAIN is less or greater than these limits, it is set to the corresponding limit. If BTAIL falls within the given bounds, but ATAIL exceeds half the value of BTAIL, then ATAIL is set to BTAIL/10.

The peak-width values determined from both reference peaks are used to determine the value of SHAPC(1) for the spectrum. SHAPC(1) is related to the "system" noise and is reevaluated for each measurement, and SHAPC(2) is related to the statistical nature of the charge-collection process in the detector. SHAPC(2) is also determined for each measurement, but its value is limited to a range of 0.0017 to 0.0019. If the calculated value of SHAPC(2) exceeds either limit, then its value is set at the corresponding limit.

The analysis of the reference peaks is repeated with these new parameter values to ensure that the approximations used in the first pass do not distort the final results. After the second pass, the values obtained by analyzing the reference peaks for the gain, zero, and FWHM of the 208-keV peak are displayed or printed. If peak areas are requested in the report, then SHAPC(1), SHAPC(2), ATAIL, and BTAIL are also displayed or printed.

Several checks are made on these parameters to verify that an accurate analysis can be completed. The gain and zero are checked to determine if the spectral regions required for the isotopic analysis are included in the spectrum. If the gain, zero, and maximum number of channels in the spectrum indicate that the maximum energy in the spectrum is less than 210 keV, the message

BAD GAIN; CHECK PEAK POSITIONS

is displayed or printed. Because the program cannot do a complete analysis if the 208-keV peak is not included, the analysis is stopped, and the program returns to the main menu. If the channel-zero energy is greater than or equal to 121.5 keV, the message

BAD ZERO; CHECK PEAK POSITIONS

is displayed or printed. Again, the data analysis is stopped, and the program returns to the main menu. If the FWHM of the 208-keV peak exceeds 1.15 keV, the message

POOR RESOLUTION

is displayed or printed. Although the analysis continues, this warns the user that the spectral resolution is not good and that the results may be questionable.

The program next examines the high-energy side of the 208-keV peak for evidence of pulse pileup. The net counts in three channels, beginning at 209.2 keV, are summed, and if the sum is greater than 0.3% of the 208-keV peak height, then the message

Significant pileup effects found in spectrum.

Results may be suspect. Check count rate and pole zero.

is displayed or printed.

The program also examines the 40- to 51-keV region (if present in the spectrum), which is immediately below the ^{241}Am 60-keV peak, for evidence of significant scattering of the 60-keV gamma ray. If the maximum of the net counts in this region is greater than half the maximum net counts of the 60-keV peak, then the message

Large scattering peaks observed in spectrum.

Analysis will be performed, but results may be suspect.

Check measurement setup. Remove objects causing scattering.

is displayed or printed. If there are objects in close proximity to the sample being measured (e.g., an object made of low-Z materials used to prop up the sample), then some of the gamma rays from the sample will scatter off the object into the detector and be counted. These are background counts, and they distort the spectrum shape, making it difficult to determine the true background. They also reduce the amount of time the system spends counting useful counts. Scattered gamma rays should be avoided by removing any objects that are near the sample being counted.

The next step in the program is to analyze seven or nine peak regions in the spectrum to determine gamma-ray peak areas. If the spectrum does not contain the 330- or 370-keV regions, only seven regions are analyzed. Each region may contain only one peak or a multiplet of overlapping peaks (peak group). The beginning and ending energies of the regions are stored in the ENSTCH and ENECH arrays. The number of channels used to determine the background levels on both sides of each region is stored in the NBG1 and NBG2 arrays.

The channel location of the region to be analyzed is determined from the gain and zero values. Once the starting channel of the region, IST, and the number of channels in the region, NDPTS, has been determined, a check is made to ensure that the entire group is contained in the spectrum. A separate check is made to determine if the upper background window overlaps the end of the spectrum. If it does overlap, the 370-keV region is not analyzed. Before the 330- and 370-keV regions are located, the gain

is reevaluated by determining the position of the ^{239}Pu 375-keV peak. This is done in case of a nonlinearity in the spectrum.

The next step is to determine the background levels on both sides of the region with the AVE subroutine, which determines the average of the counts in a specified region. The background distribution beneath the peak(s) is calculated and subtracted channel-by-channel from the region with the BKGRD subroutine. If the 203.5-keV peak area is being determined separately (i.e., the ^{235}U 185-keV peak is not present), an additional step is taken to remove any counts from the region caused by the 208-keV peak's low-energy tail. This is done with the STRIP subroutine.

After removing the background, the reference peak in each region is located with the subroutine GFIT2. For the first peak group (the 120-keV region) the position of the 129.3-keV peak is checked to ensure that the correct reference peaks were used. If the position of the 129.3-keV peak is found to be incorrect based on the gain and zero, the message

CHECK REFERENCE PEAK POSITION

is displayed, and the analysis is stopped.

The next step in the analysis of a peak or peak group in a region is to load the arrays PKHT, RATIO, and NXFLG with the information needed for fitting the peak group to determine peak areas. The byte array, STR, which contains the information for how these arrays are to be loaded, can be divided into nine parts. Each part applies to one of the nine peak groups. The first value in each part gives the number of peaks in the group. For instance, the 6 in the array (see Vol. 3) indicates that there are six peaks in the first peak group. The remaining byte values for each part indicate whether the height (area) of each peak is a free parameter or if it is fixed relative to another peak. These byte values are loaded into the NXFLG array. If a value is zero or is equal to the number of peaks in the group, the corresponding peak height is a free parameter. Any other value indicates that the corresponding peak height is ratioed relative to the free-peak height indicated by that value. For instance, in the first peak group, the value 3 for the second, third, and fourth peaks indicates that the heights of these peaks are ratioed to the height of the third free peak, which in this case, is the last peak. The corresponding ratios are obtained from the RATIO array and loaded into the appropriate locations in the PKHT array. The peak with a value in the array that is equal to the number of peaks in the group is the peak in relation to which the positions of all the other peaks are fixed.

Now, a set of linear equations is formed and fitted simultaneously to the net data channels in the group, with the peak heights as the unknowns. The calculational method is to form the matrix (peak-response matrix) of linear equations, ALPHA, and to solve it by the method of weighted least-squares. The weights, WT, are the variances of the approximate gross channel counts. The peak heights, PT, are obtained by inverting the ALPHA matrix with the MATINV subroutine and multiplying the inverted matrix by the SUM matrix. The inverted matrix elements of weighted normal equations are used for assigning errors to the results. The peak areas are determined from the peak heights by integrating the corresponding Gaussians. The area errors are determined by multiplying the square root of the chi-square value of the fit by the peak-height errors. The resulting peak energies, areas, and errors are then printed or displayed, if peak areas are requested in the report. For those peaks with heights that are fixed relative to another peak, the error value is replaced by the term FIXD in the report.

When the program determines that there are no more regions to be analyzed, it checks to make sure that an area was determined for the 208.0-keV peak. If this area is found to be zero, then the program returns to the main menu without completing the analysis.

The peak areas must be corrected for detection efficiency before isotopic ratios can be determined. These corrections require determination of the overall detection efficiency for the gamma rays emitted by the sample. A self-calibration technique is used to obtain the necessary relative

detection-efficiency corrections from peak areas in each spectrum measured. Areas of the 129.3- and 203.5-keV peaks of ^{239}Pu and the 164.6- and 208-keV peaks of ^{241}Pu are corrected for gamma-ray branching intensities, and the natural logarithms of these values are fitted to a quadratic function of the natural logarithm of the energy (in MeV). An additional coefficient is required in the fit to normalize the ^{241}Pu data points to the ^{239}Pu data points. A weighted least-squares technique is used and is done with the PUFIT subroutine. If the 267-keV peak is included in the spectrum, this ^{241}Pu peak is used, and a cubic function is used to fit the data. If the 330- and 370-keV peak groups are included in the spectrum, the 345.0- and 375.0-keV ^{239}Pu peaks are also used to determine the relative detection efficiency. In this case, a cubic function is also used to fit the data. The results of this analysis allow the relative detection efficiency to be calculated at any energy and, therefore, any peak area to be corrected.

To determine plutonium isotopic ratios, appropriate peak areas from various energy regions are incorporated into a set of linear equations to be solved by the method of least squares [see Eq. (11)]. Peak-area ratios are the dependent variables, while the isotopic-activity ratios are the independent variables. The peak-area ratios must be corrected for isotopic-branching probabilities and detection efficiencies. The equations are weighted by using (1) the peak-area errors determined in the peak-group-fitting process and (2) the relative detection efficiency for each region. If the 330- and 370-keV regions are not in the spectrum, then four equations are developed, otherwise, six. The ALPHA matrix formed by these equations is inverted by the MATINV subroutine. The plutonium-activity ratios are calculated by multiplying the inverted matrix by the BETA matrix. The errors in the ratios are obtained from the diagonal elements of the inverted matrix.

The isotopic-activity ratios are multiplied by the corresponding half-life and weight ratios to obtain the plutonium isotopic weight ratios, from which the plutonium isotopic weight-percent abundances are calculated. This calculation requires a value for the ^{242}Pu abundance, which cannot be determined by gamma-ray analysis. If the user enters a nonzero value for the ^{242}Pu abundance, then this value is used. If a zero value is entered, the ^{242}Pu abundance is calculated by means of an empirical algorithm based on the ^{240}Pu and ^{241}Pu abundances. The ^{241}Pu abundance must be corrected for decay since discharge from the reactor. Because this decay information is not always available, the ^{241}Pu abundance is corrected only for decay since separation from ^{241}Am . This period of decay can be determined from the $^{241}\text{Am}/^{241}\text{Pu}$ ratio, Z5141. The calculated age since chemical separation from ^{241}Am is used to partially correct the ^{241}Pu abundance. The corrected $^{241}\text{Pu}/^{239}\text{Pu}$ ratio, X4149, and the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio, Z4049, are used to calculate the weight-percent ^{242}Pu abundance, P242.

After calculating the weight ratios, weight-percent abundances, age since separation, and the attendant errors, these values are printed or displayed. If declared isotopic-abundance values are entered in the plutonium isotopic-abundance form, then the declared values and the percentage difference between the measured and declared values for each isotope are printed or displayed. If an analysis date for the declared plutonium and americium abundances is given in the plutonium-abundance form, then the abundances are decay corrected to the measurement date. If the analysis dates are not given, then the declared values entered should be corrected to the measurement date. After this information is printed or displayed, the program returns to the main menu.

Data-Analysis Subroutines

AVE

This subroutine determines the average value, AV, of those data values contained in the Y data array from NS to NE, inclusive. The sum of these data values is returned by the subroutine in SUM.

BKGRD

This subroutine calculates and subtracts the background distribution, channel-by-channel, from a region contained in the Y data array. NS gives the starting location, and NE the ending location of the region within the Y data array. The background levels on both sides of this region, AVBG1 and AVBG2, are provided by the calling program. The difference between these two levels is smoothed beneath the specified region and subtracted from the data in the region. (See a complete description of the background algorithm in Section 3.) The channel data, with the background subtracted, is returned by the subroutine in the YNET array. NOTE that the channel data contained in the YNET array are shifted down by NS locations from the corresponding channel data in the Y data array.

CTAIL

This subroutine removes the Gaussian component from a peak and analyzes the residual for the tail parameters, A and B. The natural logarithms of the positive residuals are fitted to a straight line by least squares to give a tail-amplitude value, A, and a tail-slope value, B. The peak must have good statistics in order to have sufficient data in the tail for analysis. (See description of peak shape in Section 3.)

DPM

This function routine calculates a pseudo relative detection-efficiency value for the isotope specified by the parameter L (^{239}Pu or ^{241}Pu) at the energy (in MeV) specified by XX. The calculation uses the polynomial coefficients determined by the log-log fit in the PUFIT subroutine. The ratio of two values at two different energies is equivalent to the ratio of the detection efficiencies at those two energies.

GFTT2

This subroutine fits the upper half of a peak with a Gaussian after removing the tailing contribution from the peak. The subroutine returns the peak position, PKPOS; the peak height, PKHT; and the peak-width parameter, ALFA. The subroutine must be provided with the position, L, of the peak within the Y data array, an approximate value for ALFA, and values for the peak-tail parameters, EXP1 and EXP2. An error condition is returned if no valid peak exists within an appropriate interval from the location, L, or if the statistics are poor. The calling program must take appropriate action.

MAXVAL

This subroutine determines the location, L, with the maximum data value, YMAX, in the Y data array.

MINVAL

This subroutine determines the location, L, with the minimum data value, YMIN, in the Y data array.

MATINV

This subroutine was adapted from the matrix-inversion subroutine given in Ref. 10. The subroutine inverts a square matrix in place and calculates its determinant. The input variables are the matrix in ARRAY and the order of the matrix, NORDER. The output is the value of the determinant, DET, and the inverted matrix in ARRAY.

PUFIT

This subroutine uses a weighted least-squares technique to fit a polynomial of order NTERMS to peak areas corrected for branching intensity and then ratioed to the 208-keV corrected peak area as a

function of the logarithm of energy (in MeV) minus the logarithm of 0.208 MeV. Selected peak areas, when corrected for gamma-ray branching probabilities, give the relative detection efficiency for the measurement at several different energies in the spectrum. Fitting these values to a polynomial gives the detection efficiency as a function of energy and allows the detection efficiency at any energy to be calculated. Peak areas from ^{239}Pu and ^{241}Pu gamma rays are used in this analysis; therefore, an additional term must be used in the fit to normalize the activity of the two isotopes.

STRIP

This subroutine is used to strip a peak or part of a peak from channel data contained in the Y array. The resulting channel data are returned by the subroutine in the Y array.

Data-Transfer Program—CICERO

A data-transfer option is provided to transfer spectral data from a Silena CICERO MCA to an IBM or IBM-compatible computer via a serial line. The CICERO MCA must be equipped with a model 8500/RSIO interface board for RS-232c communications. This board must be set to 9600 baud and data-format type A.

Data transfer is initiated by the computer, but before data transfer can occur, the CICERO must be set up for data transfer to a computer. See Vol. 2 for instructions on how to set up the CICERO MCA for data transfer to a computer.

The spectrum filename given by the user in the Clipper forms application is transmitted to the data-transfer program through a file named CICERO.PRM. The filename is then written into another file, named FILENAME.CIC, for transmittal to the data-base files.

The program first initializes the communications port in the computer to COM1. The baud rate is set to 9600, parity is set to even, databits is set to 7, and stopbits is set to 2. If an error occurs in the setup of the serial communications port, the error message,

```
Error in Setup Parameters -- Press Enter
```

is displayed.

Next, the program initiates the data transfer from the MCA with a sequence of commands. If an error occurs in issuing these commands, the following error message is displayed:

```
Error in issuing commands to analyzer
```

```
-----
```

```
The computer is unable to send commands to the  
analyzer. Check the cabling connecting the  
computer and analyzer.
```

```
Press an 'R' to retry the transfer or any other  
key to abort transfer.
```

If this error occurs, check the cable and its connections between the computer and the MCA (see Vol. 2 for cable specifications). Make sure that the cable is connected to the RS232 port on the MCA. Also check the DATA OUT option on the MCA to make sure that the MCA is set to transfer data out to a computer (see Vol. 2 for more complete instructions). If a problem is found and corrected, press 'R' to retry the data transfer.

After these housekeeping functions are completed, the program starts transmitting the spectral data in character form from the MCA, eight characters per channel. The first two characters are

control characters and are discarded. The remaining six characters are data information and are stored in a character array, DATA, for later conversion into integer values.

If no valid characters are received from the MCA, the following message is displayed:

Error in receiving data from the analyzer.

Make sure that the analyzer is connected properly and is in the correct mode for transfer.

If the analyzer is connected properly, and is in the correct mode, then the analyzer cannot detect the commands coming from the computer. You will need to manually set the analyzer to send data out to the computer.

Enter an 'M' to manually set the analyzer or a 'R' to retry automatic transfer or any other key to abort and return to the main menu

If this error message is displayed, check the MCA DATA OUT setup and the cabling and then press 'R' to retry the transfer. If the error message is displayed again, try transferring the data manually. To do this, start the data transfer on the computer, then immediately press the DATA OUT pushbutton on the MCA. This will manually transmit the data from the MCA to the computer.

If the program receives valid characters from the MCA, the following messages are displayed.

Estimated transfer time: 40 Seconds.

Now receiving data. . .

Forty seconds, the time needed to transfer a 4096-channel spectrum, is the upper limit for the amount of time required. Spectra with fewer channels will take less time, but the program cannot determine, in advance of the transfer, the number of channels that will be transferred.

Once the data transfer is complete, the number of channels transferred will be displayed as follows:

Transfer completed with <#> channels sent

Now writing data file.

If the number of channels transferred is greater than 4100, then the message

Only 4100 channels recorded.

is displayed. The program can accommodate only 4100 channels, so if more than 4100 channels are transmitted, only 4100 channels will be stored in the spectral data file.

This is followed with the messages

Spectrum ID: <#>

Livetime: <#>

which give the spectrum ID number assigned by the CICERO MCA and the spectrum livetime.

The program now converts the ASCII-character data into I⁴ values and writes them into the binary spectral data file. The program also checks the data for any zero counts. If a channel is found to have zero counts and its neighboring channels to have nonzero values, then the following message is displayed:

WARNING

Zero count channel found at channel number <#>

This spectra file may not be usable.

Retry data transfer.

Press Enter to Continue

A channel with a zero-count value may indicate a "dropped" channel in the MCA or an improper transfer of the data from the MCA. Check the spectral data in the MCA display to determine whether a zero-count value for the identified channel is appropriate.

Once the spectral data file has been written to disk, the message

Press Enter to continue.

is displayed, and doing so will return the program to the main menu.

Data Quality-Assurance Program—DCHECK

DCHECK is a data-analysis option that checks certain parameters needed for the system setup and performance of a plutonium isotopic analysis. The gain, zero, and peak-shape parameters (i.e., resolution, tailing, and pileup) are determined by analyzing low- and high-energy peaks in a gamma-ray spectrum from any gamma-ray emitter (i.e., it does not have to be a plutonium gamma-ray spectrum).

Inputs to the program are the energies and channel positions of the low- and high-energy gamma-ray peaks and the spectrum filename. The two peaks should be well isolated (free from interferences) and should contain a minimum of 10^5 counts (or a peak height of 2×10^4 counts) to ensure a good determination of the gamma-ray peak shape.

The program locates the peaks indicated and analyzes them for approximate positions and FWHM values. These parameters are determined by using the GFIT2 subroutine to fit the tops of the peaks indicated. If the measured width parameter is positive (instead of the expected negative value), the following error message is displayed:

Error - Bad Shape Parameter

This error may be the result of an incorrect entry for the peak position or poor statistics for the peak indicated. Check the spectrum and take appropriate action.

If the peak-width parameters are negative, the program proceeds by removing the backgrounds from beneath the peaks with the NCNTS subroutine. It then fits the peak tops again with the GFIT2 subroutine for more-precise peak positions and width parameters. If an error occurs in GFIT2 when fitting the peak tops, the following message is displayed:

BAD PEAK POSITION

Check the spectral data for the correct positions of the peaks and for any possible interferences with the peaks being analyzed. If there is another peak within 10 channels of the peak being analyzed, this error could occur.

If an error is not returned from GFIT2, the program calculates the gain and zero for the system and the peak-shape parameters SHAPC(1) and SHAPC(2), which are the K_1 and K_2 terms in Eq. (5). The program then checks the high-energy side of the high-energy peak for evidence of pileup. If the sum of the net counts in the three channels starting 1.2 keV above the peak centroid is greater than 0.3% of the peak height, then the program will indicate the presence of pileup.

The program now displays the results of the analysis along with any appropriate diagnostic messages. First, the spectrum lifetime and the count rate obtained from the spectrum are displayed. Next, the values obtained for the gain and zero are displayed. If the gain is greater than 0.15, the message

Gain MUST BE LESS than 0.15 keV/channel

is displayed. This requirement exists to ensure that sufficient channel spacing exists between gamma-ray peaks for a plutonium isotopic analysis. Compression of the spectrum by means of a high gain makes it difficult to obtain an accurate background level in some regions of the plutonium spectrum.

If $GAIN * (\text{maximum channels in spectrum}) + ZERO \leq 210 \text{ keV}$, the message

CHECK GAIN; Upper energy range MUST BE AT LEAST 210 keV

is displayed. The 208-keV peak must be present in a plutonium gamma-ray spectrum for an analysis to be done. Adjust the gain and the zero-channel offset to ensure this.

If the channel-zero energy is greater than 121.5 keV, the message

CHECK ZERO; Zero CANNOT BE GREATER than 121.5 keV

is displayed. The 120-keV region must be present in a plutonium gamma-ray spectrum for an analysis to be done. Check the zero-channel offset to ensure that the zero-channel energy is less than 121.5 keV.

Next, the program displays the calculated resolution values at 208 and 122 keV. The resolution is calculated at these two energies by using the resolutions determined at the two reference peaks. If the resolution calculated at 208 keV is greater than 1.15 keV, then the message

POOR RESOLUTION -- CHECK SYSTEM AND COUNT RATE

is displayed. Reduce the count rate, if possible, or check the amplifier pole-zero adjustment to improve system resolution.

Finally, if the program detects evidence of pulse pileup in the spectrum, the message

HIGH-ENERGY TAILING IS SIGNIFICANT

CHECK SYSTEM POLE-ZERO SETTING AND INPUT COUNT RATE

is displayed. Pulse pileup should be avoided, because it distorts the gamma-ray peak shape and causes inaccuracies in determining peak areas in regions where peaks overlap.

4. Error Messages

Several checks are incorporated into the software to detect situations that might cause errors or poor results. If certain conditions are not met by the spectral data or the information entered by the user, the following error messages are printed or displayed. A brief description is also given for the meaning of each error message.

Zero count channel found at channel number <#>.
Aborting analysis. Transfer data again and retry
analysis with new spectral data.

Channels with zero counts were found in the spectrum. This may be a result of "dropped" channels in the analyzer (bad memory in the analyzer) or a problem with the transfer of data from the analyzer to the computer. Because these "bad" channels may give erroneous results, the analysis is not performed. Check the data in the analyzer; if it is okay, then transfer the data again, making sure that the transfer is being done properly.

Required array size exceeds dimension.
Increase gain.

The data-array dimension in the program is 210. If the spectrum is taken at a gain that results in an energy interval exceeding 210 data points, or if the gain is less than 0.05 keV/channel, this error message will occur.

BAD PEAK POSITION FOR REFERENCE PEAK

If statistically significant data do not exist near the peak positions indicated by the user in response to the peak-position queries, this error message is displayed or printed. This indicates that an incorrect value was entered for a reference peak position.

CHECK REFERENCE PEAK POSITION

This error condition is displayed or printed if one or more incorrect reference peaks are used. The calculated gain and zero may satisfy the necessary conditions and will be printed out; however, the program checks to make sure that the 129.3-keV peak is found in the correct position. Instead of continuing, the program returns to the main menu for the user to take corrective action and restart the analysis.

BAD PEAK-WIDTH PARAMETER; CHANNEL OVERFLOW?

An error may occur in the determination of the FWHM parameter as a function of energy. If the peak-shape analysis results in unrealistic values, this message is displayed or printed. This may occur if the calibration peaks, 148.6- and 208-keV, have very poor counting statistics, or if incorrect peak positions were entered, or if either peak has a channel that has overflowed. This condition is most likely to occur for the 208-keV peak and results in a distorted peak shape. The overflow cannot be corrected, and the user must guard against it because such a spectrum cannot be analyzed. REMEMBER: the Silena CICERO analyzer can accommodate only 999,999 counts per channel!

BAD ZERO; CHECK PEAK POSITIONS

If statistically significant peaks are found, the software proceeds to determine more-accurate peak positions from which the gain and channel-zero energy of the spectrum are calculated. If the channel-zero energy is greater than 121.5 keV, this message is displayed or printed. This indicates that the channel-zero energy precludes analysis of the 120-keV peak group, which is required by the program. This condition may exist because of an incorrect peak-position entry or the improper energy calibration of the amplifier/analyzer.

BAD GAIN; CHECK PEAK POSITIONS

If the product of the spectrum gain and maximum number of channels in the spectrum (4096) plus the zero-channel energy is less than 210 keV, this message is displayed or printed. The program cannot analyze the spectrum if the 208-keV peak region is not included. Again, either the correct peak position must be entered, or the spectrum must be retaken with a proper energy calibration of the amplifier/analyzer.

POOR RESOLUTION

If the FWHM of the 208-keV peak exceeds 1.15 keV, this warning message is displayed or printed to warn the user that the spectral resolution is not good and that the results may be questionable. Although the data analysis will continue, the user should improve the energy resolution by simply reducing the count rate or by adjusting the amplifier pole-zero and shaping-time constant [consult the Instruction Manual for Instrumentation (IMI) for more details]. Energy resolution of 1.15 keV for the 208-keV peak is a reasonable value for a large coaxial germanium detector; however, for smaller planar detectors, 0.8 keV or less can be obtained. Because the software is written for all types of germanium detectors, an upper limit of 1.15 keV for energy resolution at 208 keV is used for all spectra.

Significant pileup effects found in spectrum.

Results may be suspect. Check count rate and pole zero.

The program examines the high-energy side of the 208-keV peak for evidence of pulse pileup. The net counts in three channels beginning at 209.2 keV are summed, and if the sum is greater than 0.3% of the 208-keV peak height, this message is displayed or printed. Reduce the count rate or adjust the pole zero to reduce pileup.

Large scattering peaks observed in spectrum.

Analysis will be performed, but results may be suspect.

Check measurement setup. Remove objects causing scattering.

The program examines the 40- to 51-keV region of the spectrum (if present in the spectrum), which is immediately below the ^{241}Am 60-keV peak, for evidence of significant scattering of the 60-keV gamma ray. If the maximum of the net counts in this region is greater than half the maximum net counts of the 60-keV peak, then this message is printed or displayed. If there are objects near the sample being measured (e.g., an object made of low-Z materials used to prop up the sample), then some of the gamma rays from the sample will scatter off the object into the detector and be counted. These are background counts, and they distort the spectrum shape, making it difficult to determine the true background. They also reduce the amount of time the system spends counting useful counts. Scattered gamma rays should be avoided by removing any objects that are near the sample being counted.

BAD 375.0 keV PEAK

If the 330- and 370-keV regions are in the spectrum being analyzed, then the 375.0-keV peak is used as an additional reference peak to adjust the gain and channel-zero energy of the spectrum in case nonlinearity exists. The location of the 375.0-keV peak is determined with the GFIT subroutine that fits the top of the peak. If a problem occurs in this process, this error message will be printed or displayed. A problem would occur if the 375.0-keV peak has poor statistics or a very poor peak shape.

208-keV peak area is ≤ 0 . Analysis Aborted.

The 208-keV peak must exist for a complete analysis. The 208-keV peak is essential for determining the relative-efficiency curve.

<energy>-keV peak area is ≤ 0 . Analysis is aborted!

One of the peak areas used in establishing the relative-efficiency curve was found to be negative or zero. Consequently, insufficient information exists to determine the relative-efficiency curve, and the analysis is aborted. This condition is likely to occur if the data have very poor statistics.

Pu-240 160-keV area = 0. Results are suspect.

The ^{240}Pu 160-keV peak is the only peak above 120 keV that is available for determining the ^{240}Pu abundance. If the area of this peak is found to be zero, then the ^{240}Pu abundance cannot be determined, and the results are incorrect. This condition is probably caused by very poor statistical data or by problems with the analysis of the 160-keV region.

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