CONF-9007106--73

DE90 017824

SPREADSHEET ANALYSIS OF GAMMA SPECTRA FOR NUCLEAR MATERIAL MEASUREMENTS

by

W. R. Mosby and D. M. Pace*

Special Materials Division
Argonne National Laboratory
P. O. Box 2528
Idaho Falls, Idaho
83403-2528

DISCLAIMER

For presentation at the
INMM 31st Annual Meeting
July 15-18, 1990
Los Angeles, California

*Work supported by the U.S. Department of Energy, Civilian Reactor Development, under Contract W-31-109-ENG-38.

Abstract

A widely available commercial spreadsheet package for personal computers is used to calculate gamma spectra peak areas using both region of interest and peak fitting methods. The gamma peak areas obtained are used for uranium enrichment assays and for isotopic analyses of mixtures of transuranics. The use of spreadsheet software with an internal processing language allows automation of routine analysis procedures increasing ease of use and reducing processing errors while providing great flexibility in addressing unusual measurement problems.

Introduction

At Argonne National Laboratory-West (ANL-West), gamma spectroscopy based nondestructive assay (NDA) is usually accomplished in a two part process as shown in Figure 1. First, specially written interactive software running on a computer at a measurement location is used to control gamma-ray detection and measurement equipment and to store gamma-ray spectrum data on a floppy disk. After the data is collected, a widely available spreadsheet software package, Microsoft Excel™, is used to access the stored spectrum data and perform assay calculations for both routine and unusual assays. The calculations may be done on the measurement computer or on another computer in the NDA laboratory.

This two part process arose naturally at ANL-West. Software written to interface with specific measurement hardware is not often changed because hardware changes rarely occur. On the other hand, the NDA organization at ANL-West is required to perform many different routine and specialized gamma-ray measurements. This requires a flexible analysis software system.

This paper describes the routine and specialized analysis spreadsheet systems in use at ANL-West. The spreadsheet development process is presented. Data processing and pertinent calculations performed by the analysis are discussed.

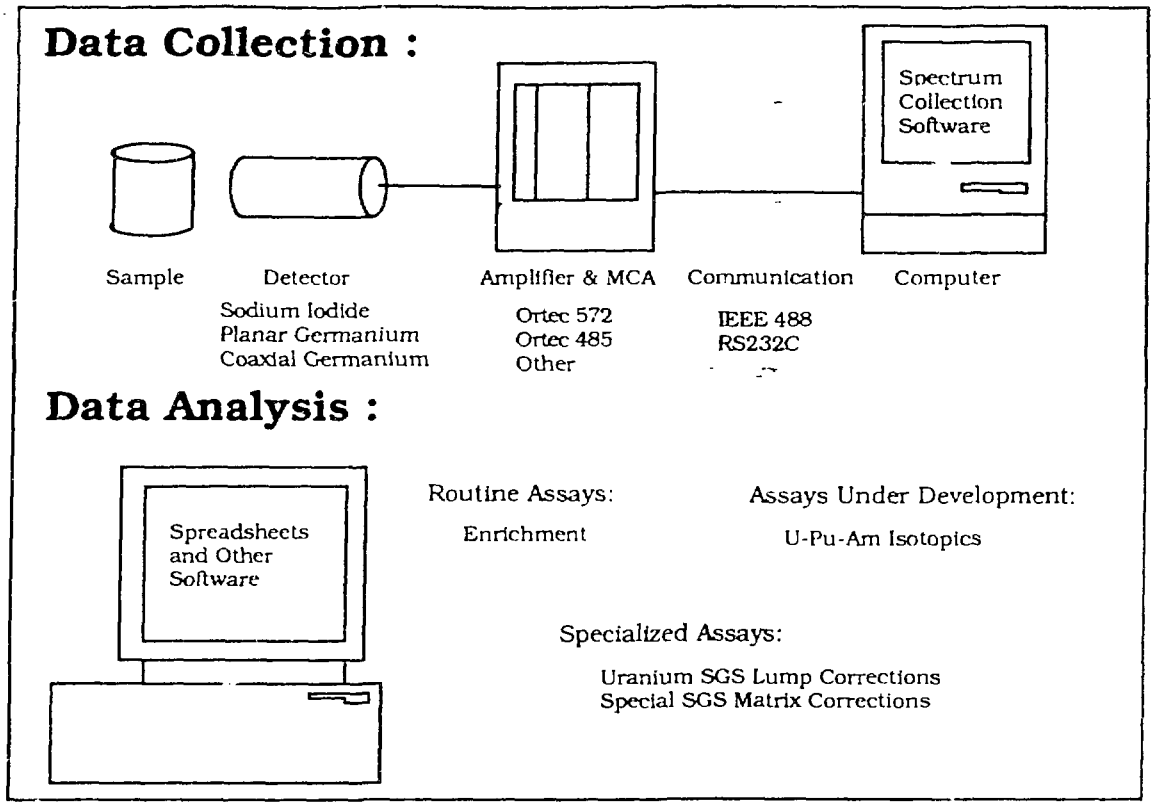

Figure 1: NDA Gamma Measurement Process at ANL-West

Spreadsheet Development

Microsoft Excel has been found to be very useful in conceptualizing, developing, and applying gamma-ray assay analysis methods to changing measurement requirements. The process begins with the creation of a spreadsheet containing relevant data input, processing, and results regions. The spreadsheet may be accompanied by one or more auxiliary spreadsheets and "macro" spreadsheets. A macro is a program written using the functions provided in Excel. Macros provide control of the data processing. As development proceeds, components of the software system are modified until a finished, documented analysis system results. Figure 2 summarizes the spreadsheet development process.

Analysis Spreadsheet Development Process:

Basic formulas entered into preliminary spreadsheet, basic tests performed.

Simple macros written to read one data set at a time into the spreadsheet for further tests.

Analysis formulas moved from spreadsheet into a more comprehensive macro.

Auxiliary spreadsheets created and used for standard data, background calculations, tamper indicating device ("TID") records, etc.

Analysis spreadsheet becomes a repository for data and results.

Macros developed to be as fully automated as possible.

Finished assay spreadsheet shows item ID, TID, book values, count rate data, NDA results, comparison to book values, macro and spreadsheet version numbers, etc.

Figure 2: The Analysis Spreadsheet Development Process

Hardware Interface and data collection software

Apple™ Macintosh™ computers are used for data acquisition and analysis, as well as for assay results reporting. The PCs in use in the ANL-West NDA lab range from the Macintosh Plus to the Macintosh IICX.

Various germanium and sodium iodide detectors are used at ANL-West, along with a number of different amplifiers. The EG&G Ortec 918A multichannel analyzer (MCA) is interfaced to the Macintosh using either RS232C or IEEE 488 interfaces. This is a microprocessor-based fixed conversion time MCA with a spectrum storage capacity of 8192 channels of 24 bit word length.

All Macintosh models have built-in serial ports which allow communication with the MCA using the RS232C interface. A Macintosh SE incorporating an IEEE 488 interface card uses this byte parallel protocol to rapidly transfer spectrum data. Transfer speed is especially important during setup of 8192 channel spectra.

The data collection software, called "MacMCA", is a program developed by the NDA Laboratory at ANL-West that supports the Macintosh user interface. MacMCA acquires and displays gamma spectra. The user can elect to store spectrum data at three levels of detail: the full spectrum, the spectrum details in each region of interest (ROI), or the first channel count, last channel count, and total count for each ROI. In addition, MacMCA allows the user to write an ASCII text copy of this stored data to disk for the use of analysis software. Figure 3 presents an example of a gamma spectrum display from MacMCA. It shows the low energy region of an enriched uranium spectrum taken with a sodium iodide detector.

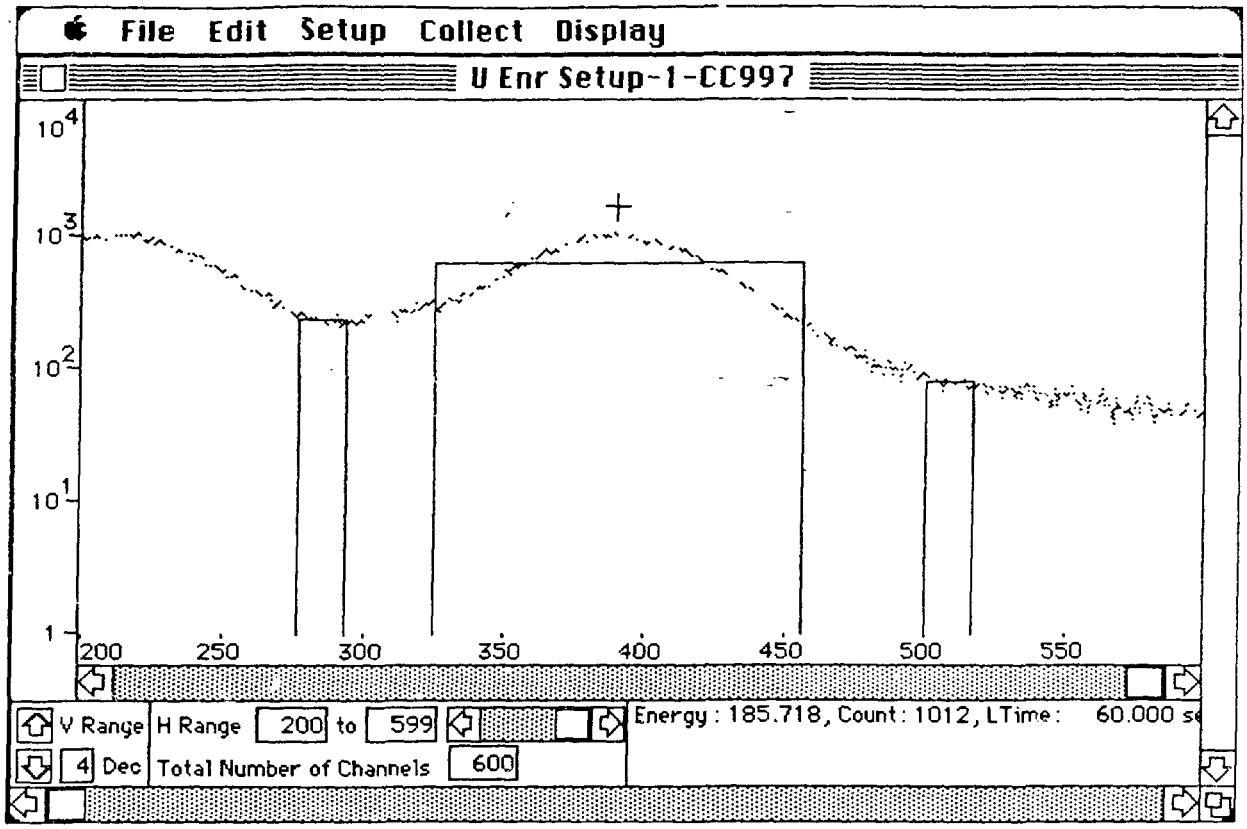

Figure 3: MacMCA Screen Display

Routine Enrichment Analysis

Region of Interest (ROI) peak area calculations determine the U-235 enrichment of uranium bearing metal alloys. For the enrichment assay, MacMCA is used to acquire the gamma count data from a sodium iodide measurement system. MacMCA is set up to produce a file containing lower and upper background ROIs and an ROI for the 186 kev peak of U-235.

In the text file written by MacMCA, data are separated with tabs and carriage returns, facilitating the use of this data by a spreadsheet. Microsoft Excel allows a text file to be opened as a spreadsheet. Each tab-separated field in a line is placed into a spreadsheet column, while each carriage return starts a new row in the spreadsheet.

The "Enrich" macro controls the analysis process. This macro operates on four files: the count data file, an optional item book data file, an enrichment standard data file called "Enrich Stds", and an assay report form called "Enrich Results". The macro extracts ROI data, count time, book values, and other relevant data, performs computations, reports assay results, and performs other essential functions associated with an NDA measurement. Figure 4 shows a schematic diagram of the Enrich spreadsheet system.

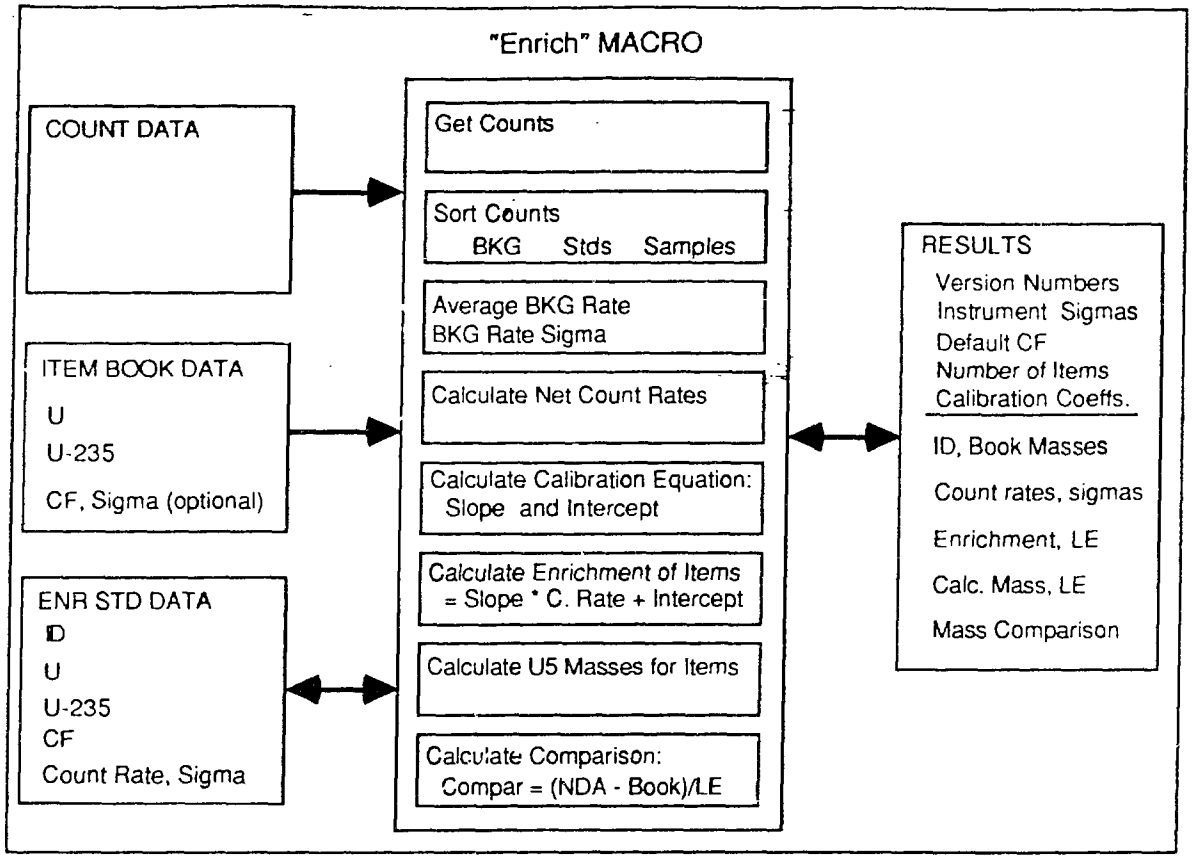

Figure 4: "Enrich" Spreadsheet System

To begin the processing, Enrich opens the text file written by MacMCA which causes Excel to place the data in a spreadsheet, generating the count data file. Enrich sorts background, enrichment standard, and sample count data into appropriate data storage spreadsheet areas.

After sorting and storing all the count data, Enrich determines the net peak areas for the standard and samples, propagating the count uncertainty. Enrich calculates the net area of a peak by subtracting a linear background profile [1] from the total count in the peak ROI. The linear background profile, shown in figure 5, is determined by interpolation using lower and upper background ROIs. Enrich also applies a default or user-supplied container attenuation correction to the enrichment standard and measured item count rates.

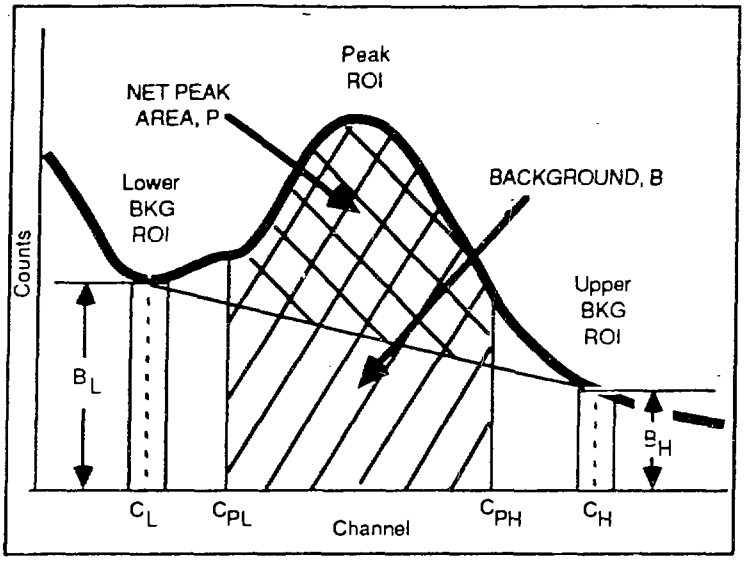

Figure 5: "Enrich" Net Peak Count Calculation Schematic

The formulas used by Enrich to calculate the peak area from the background and peak ROIs are given below.

The net area of the 186 kev peak, P, is given by:

$$P = T - B, \qquad (1)$$

where T is the total count in 186 kev peak region of interest (ROI) and B is the continuum background under the 186 kev peak.

The continuum background may be calculated from the formula:

$$B = \frac{\left[\frac{B_H-B_L}{C_H-C_L}\right]\left[C_{PL}+C_{PH}\right]+2\left[B_L - C_L\left[\frac{B_H-B_L}{C_H-C_L}\right]\right]}{2} \times$$

$$\times \left[C_{PH}-C_{PL}+1\right] \qquad (2)$$

where B_L and B_H are the average counts per channel in the lower and higher background ROIs, C_L and C_H are the middle channels of the lower and higher background ROIs, and C_{PL} and C_{PH} are the low and high channels of the peak ROI.

The uncertainty of the net area of the 186 kev peak is given by:

$$\sigma_P = \sqrt{\sigma_T^2 + \sigma_B^2} \quad , \qquad (3)$$

where $\sigma_T = \sqrt{T}$ is the standard deviation of the total peak area and σ_B is the standard deviation of the linearly interpolated background, B. The following formulas give the standard deviation of the background, σ_B:

$$\sigma_B{}^2 = \left[\frac{\partial B}{\partial B_L}\,\sigma_{B_L}\right]^2 + \left[\frac{\partial B}{\partial B_H}\,\sigma_{B_H}\right]^2 \qquad (4)$$

Expanding,

$$\sigma_B{}^2 = \left[\frac{C_{PH}-C_{PL}+1}{C_H-C_L}\right]^2\left[\left\{\left(C_L-\frac{C_{PH}+C_{PL}}{2}\right)\sigma_{B_L}\right\}^2 + \left\{\left(\frac{C_{PH}+C_{PL}}{2}-C_L\right)\sigma_{B_H}\right\}^2\right] \qquad (5)$$

where σ_{B_L} and σ_{B_H} are the standard deviations of the average counts per channel in the lower and upper background ROIs, respectively. σ_{B_L} and σ_{B_H} may be calculated from: $\sigma_B = (\sqrt{\text{BKG ROI count}}) \div (\text{number of channels in BKG ROI})$ for each background region.

After calculating the peak areas and uncertainties, Enrich calculates the coefficients of the enrichment calibration function, which gives enrichment as a linear function of net count rate. The calibration function coefficients are determined using a least squares fit [2] to the count rate vs enrichment data for the standards used in the assay. One or more standards may be used. The calibration coefficients and the net count rate for each item is used to calculate each item's enrichment and the limit of error (L.E. $\equiv 2\,\sigma$) of the enrichment.

Finally, if a book value file is present, Enrich uses the uranium mass given for each item and the measured item enrichment to calculate the U-235 mass and L.E. for the item. A calculation is performed to compare the measured item and L.E. with the book value.

The results spreadsheet contains book value, NDA, and comparison data for each measured sample as well as calibration parameters, macro and spreadsheet version numbers, instrument variance, balance limit of error used, and the default item attenuation correction factor.

Enrich provides an automated means of analyzing ROI data. After opening the Enrich macro from the Macintosh desktop display, the user is prompted for the name of the file containing the count data, for the book value file, if any, and for the limit of error of the balance used in weighing the measured items. The macro then finishes the analysis and produces a results spreadsheet.

The process from data collection to report generation is performed by measurement personnel; the data and report printouts are reviewed by analysis personnel. This automated system minimizes operator errors.

Developmental Isotopic Ratio Analysis

The peak fitting process described is an example of an assay analysis system under development. The data acquisition software, MacMCA, is used to acquire a high-resolution spectrum of a measured item containing some combination of uranium, plutonium, americium, and possibly neptunium. MacMCA is set up to write all the channel count values in the full spectrum to disk. A system of

macros and spreadsheets is used to extract the spectrum data and perform curve fits. Figure 6 shows a schematic of the macros and spreadsheets used.

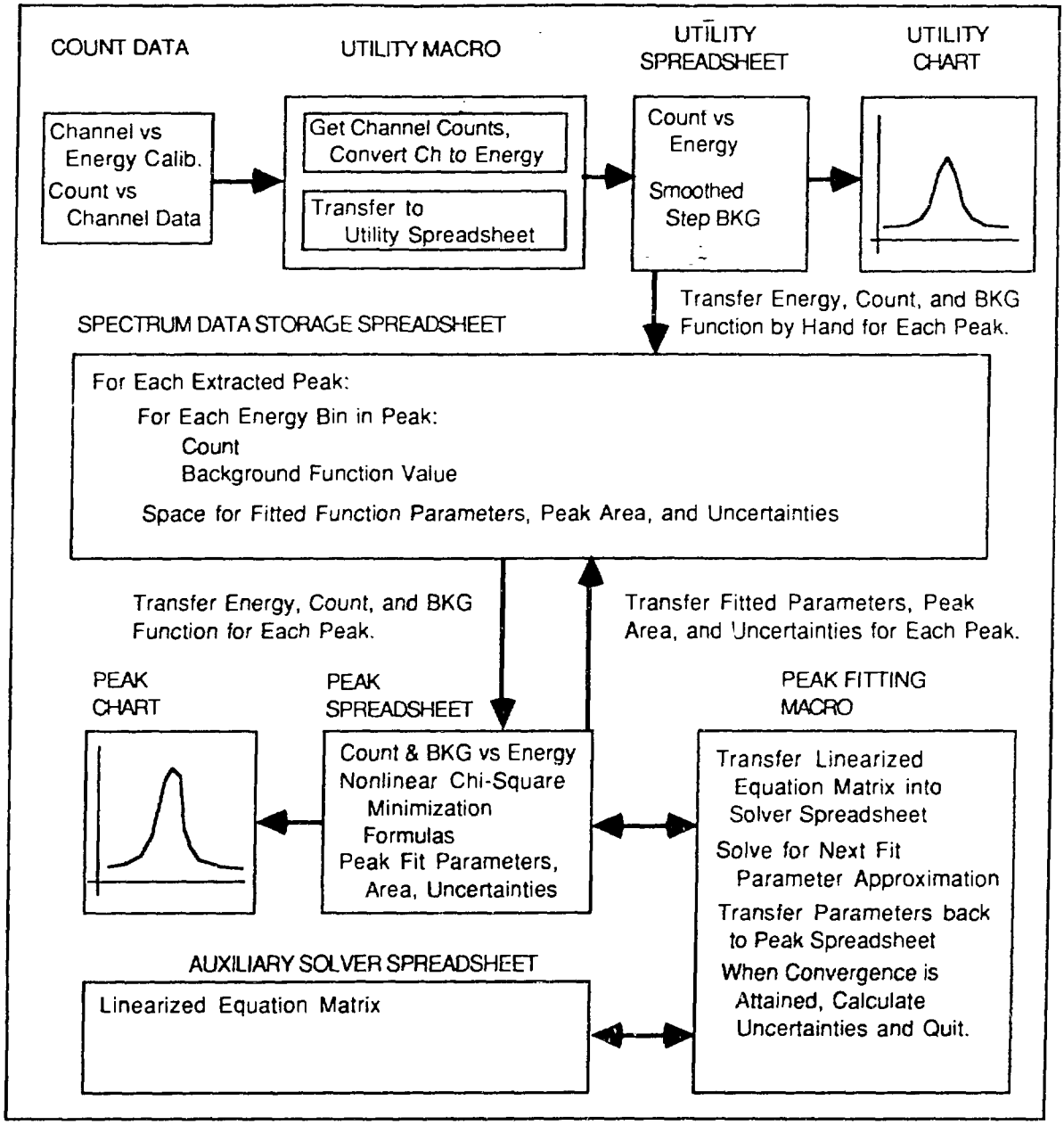

Figure 6: Peak Fit System

Utility macro, spreadsheet, and chart documents are used to extract count data for each peak of interest in the spectrum. The spreadsheet and chart are linked to give a graphic view of the extracted peak. The spreadsheet also calculates a smoothed step background function [1] which is used in the peak fitting process. This background function is of the form:

$$B_i = \sum_{j=i}^{n} C_i \Big/ \sum_{j=0}^{n} C_i, \qquad (6)$$

where C_i is the count in channel i. This formula is applied only to the region surrounding a peak rather than to the entire spectrum. The extracted peak energy, count, and background data are pasted into a spreadsheet used for spectrum data storage. All peaks of interest are extracted from the spectrum and stored in this peak data spreadsheet.

The data extraction spreadsheets are closed and a set of peak fit spreadsheets are opened. This set consists of a peak calculation spreadsheet, a macro sheet, a linear equation solver spreadsheet, and a peak chart sheet. The peak chart sheet shows a plot of the fitting function, the MCA data points, and the error between the two. This allows the user to check the initial function parameters and to follow the progress of the peak fit algorithm. Figure 7 below shows a screen display of the peak chart.

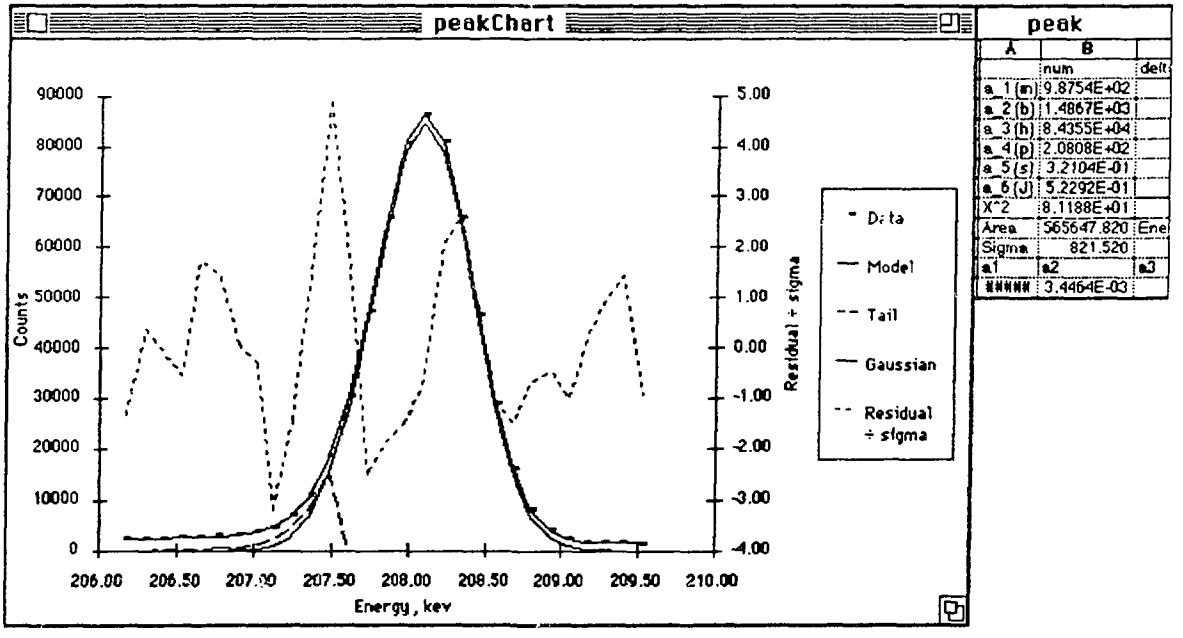

Figure 7: Peak Fit Spreadsheet System

The user performs a peak fit by transferring a peak data set into the peak calculation spreadsheet, adjusting the peak formula region to conform to the length of the data set, entering the initial peak parameters into the calculation spreadsheet, and then running the macro. The user has complete control over the fitting process. The user can set the convergence criterion, allow the fit to include a low energy tailing function, and stop the fitting macro at any time.

For a single gaussian peak with a low energy tail and a background function of the form given in equation (6), a peak fitting function of the following form may be used [1].

$$y(x_i;\mathbf{a}) = a_1 B_i + a_2 + P(x_i;a_3, a_4, a_5, a_6), \qquad (7)$$

In this equation, $y(x_i;\mathbf{a})$ denotes the count as a function of discrete energy x_i and function parameter vector $\mathbf{a}.= [a_1, a_2, a_3, a_4, a_5, a_6]$. The parameters a_1 and a_2 are used adjust the normalized background function. a_3 is the magnitude of the gaussian peak or exponential or exponential tail. a_4 and a_5 are the centroid and width of the gaussian, respectively. a_6 is the distance (in energy units) from the gaussian centroid at which the low energy tail starts. B_i is the smoothed step background function at energy i, and P is the peak function. P is of the form [1]:

$$P = \begin{cases} a_3\, e^{\,a_6(2x_i-2a_4+a_6)/(2a_5^2)} & \text{for } x_i<a_4-a_6 \\ a_3\, e^{\,-(x_i-a_4)/(2a_5^2)} & \text{for } x_i\geq a_4-a_6 \end{cases} \quad . \qquad (8)$$

The first half of equation (8) gives the low energy exponential tail function, while the last half gives the gaussian peak function. For moderate count rates, we have found that the exponential tail contributes only a few percent to the peak area. See Figure 7.

The peak fitting macro uses a nonlinear chi-square minimization algorithm [3]. The basic iteration formulas are:

$$\left[\ \alpha(\mathbf{a}_i)\ \right]\left(\delta\mathbf{a}_i\right) = \left(\beta(\mathbf{a}_i)\right) \quad \text{and} \qquad (9)$$

$$\left(\mathbf{a}_{i+1}\right) = \left(\mathbf{a}_i\right) + \left(\delta\mathbf{a}_i\right) \quad . \qquad (10)$$

Each term in equation (9) is a function of $\mathbf{a}$, = $[a_1, a_2, a_3, a_4, a_5, a_6]$, the vector of parameters of the function used in fitting the peak data. To solve for the parameters giving the best fit, an initial estimate of the parameters is made, allowing the terms of equation (9) to be calculated. Equation (9) is then solved for $\delta\mathbf{a}$, and equation (10) is used to calculate a new, better estimate of the fitting parameters. The terms of (9) are recalculated using the new $\mathbf{a}$, and the process is repeated until $\delta\mathbf{a}$ becomes as small as desired, at which time the parameters $\mathbf{a}$ may be used to calculate the fitted peak area.

The elements of the $[\alpha]$ matrix and the (β) vector are given by the following formulas.

$$\alpha_{i,j} = \sum_{k=1}^{n} \frac{1}{\sigma_k^2}\left[\frac{\partial y(x_k;\mathbf{a})}{\partial a_i}\,\frac{\partial y(x_k;\mathbf{a})}{\partial a_j}\right] \qquad (11)$$

$$\beta_j = \sum_{k=1}^{n} \frac{1}{\sigma_k^2}\left[y_k - y(x_k;\mathbf{a})\right]\left[\frac{\partial y(x_k;\mathbf{a})}{\partial a_j}\right] \qquad (12)$$

where y_k is the count in the measured gamma spectrum at energy k, $y(x_k;\mathbf{a})$ is the peak fitting function, which is a function of the energy, x_k, and a number of function parameters represented by the vector **a** and n is the number of (energy, count) pairs. σ_k is the standard deviation of y_k, and is calculated from the formula $\sigma_k = \sqrt{y_k}$.

The covariance matrix of **a** is found by computing $[C] = [\alpha]^{-1}$. The variance of a function f(**a**) may be computed from the following formula.

$$\sigma_f^2 = \sum_{i=1}^{n} \frac{\partial f}{\partial a_i} C_{ii}^2 + 2 \sum_{i=1}^{n} \sum_{j=i+1}^{n} \frac{\partial f}{\partial a_i} \frac{\partial f}{\partial a_j} C_{ij} \quad (13)$$

When the the macro is finished, the user transfers the peak fit parameters and the peak area and the uncertainties for these quantities from the calculation spreadsheet to the peak data spreadsheet.

Similar spreadsheets have been written for fitting up to three peaks. These peak separation spreadsheets require peak width as input data and fit gaussian functions of known width to the peaks. Peak width as a function of energy is determined from fits on singlet peaks in the spectrum under analysis.

The peak fitting system has been found useful for separating unresolved multiplet peaks required for measuring isotopic ratios in U-Pu-Am alloys. A separate spreadsheet system is used to calculate the isotopic ratios using the fitted peak areas as input.

An example of a doublet peak separation is shown in Figure 8. The data for this figure was taken from a sample of plutonium metal containing 23.5% Am-241 by weight. The doublet consisted of the 375 kev Pu-239 peak and the 376.6 kev Am-241 peak. The Am-241 to Pu-239 weight ratio was calculated from the separated peak area ratio, the isotope half lives, and the gamma-ray branching ratios. The calculated ratio was .22 ± .06 (2 σ), which is statistically equivalent to the book ratio for the sample.

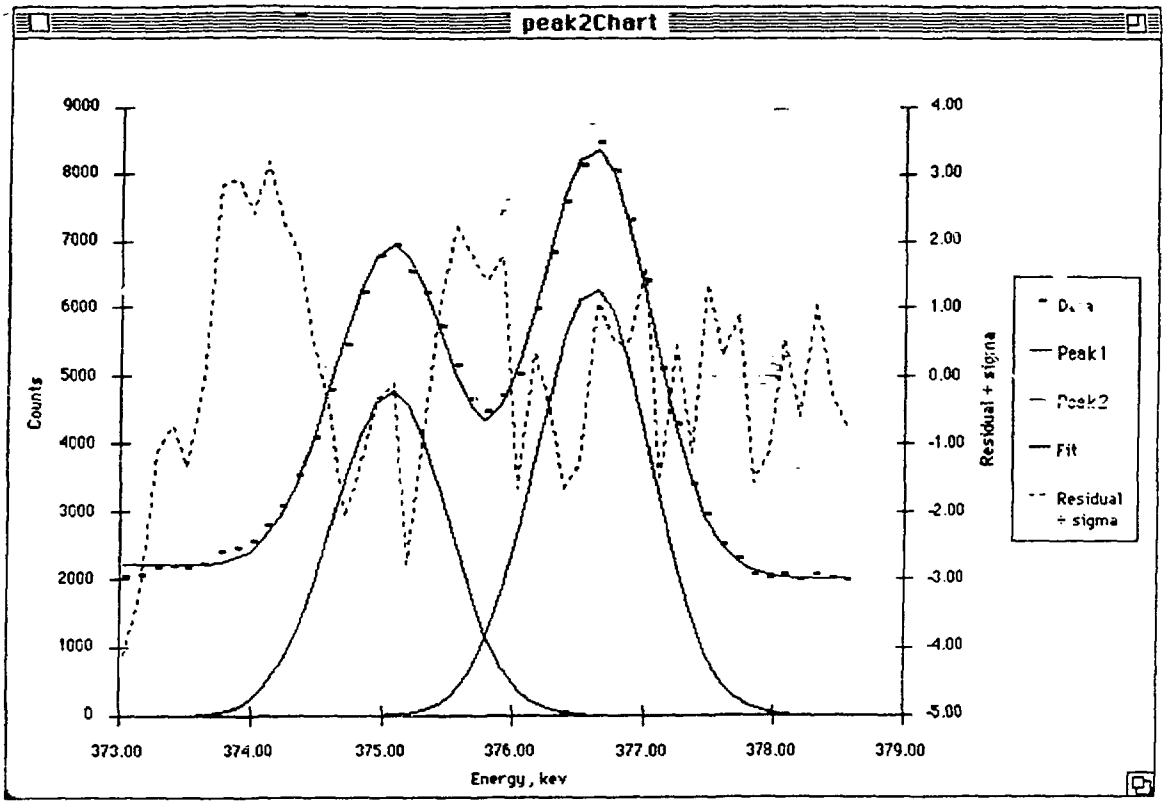

Figure 8: Doublet From Sample Containing Am-241 and Pu-239

This spreadsheet system has been very useful in developing the peak fit algorithms, and the techniques perfected during its use will be incorporated into the MacMCA spectrum acquisition system in the future.

Examples of modifications / additional calculations applied

A number of modified versions of standard assay calculations have been developed and used in support of unusual measurement requirements at ANL-West. In addition, spreadsheets have been developed which complement standard assay calculations to allow them to be used in unusual situations. Examples of these modifications and corrections are given below.

Glass scrap mixed with uranium fines and dust is routinely assayed using segmented gamma scanning (SGS). For some such items, the dust may aggregate into clumps which are self-shielding, or the sample may contain particles of uranium large enough to self-shield significantly. A macro and spreadsheet system was developed which used counts from the 143 kev and 186 kev U-235 lines to calculate a lump correction factor.

A spreadsheet has been developed which calculates the matrix correction factor for an SGS measurement. The spreadsheet performs a numerical integration which calculates the attenuation of gamma rays by an emitting absorber with a circular cross-section. The spreadsheet's input consists of the attenuation of gamma rays directed through the diameter of the circular cross-section, the cross-section radius, and the distance from the cross-section center to the detector. The spreadsheet's output is the matrix correction

factor. This spreadsheet is used for items whose combination of density and radius makes the approximate matrix attenuation correction used by the lab's standard SGS software inaccurate [4].

A modified version of the "Enrich" macro described above has been developed. This version takes an item's count and count standard deviation directly as input instead of calculating it from background and peak ROI information. This version of Enrich can be used to process data from an Eberline Instrument Corporation Stabilized Assay Meter (SAM-2) enrichment measurement system.

These spreadsheet systems were developed in short times, ranging from hours to a few days. This illustrates one great advantage of spreadsheet software development-- its flexibility and transparency. The results of calculations are always displayed and changes and corrections can be made rapidly as the analysis develops.

Conclusions

There are many advantages to spreadsheet systems. They are largely self-documenting. Dialog boxes, custom menus, and user messages can be used to make the resulting software user-friendly. It is easy to generate computer screen shots of the software in operation for inclusion in illustrated manuals and procedure documents. Spreadsheet formulas and constants may be printed, providing documentation of the formulas and algorithms used.

There are a few disadvantages of a system using separate data collection and data analysis software. Data analysis is done off-line, after the measurement data is taken. However, control measurement data reduction must be performed at the time of the assay to ensure that valid measurement data is taken. In practice, the data reduction required has been found to be easily done by task switching the computer between the data acquisition program and a spreadsheet used to perform the control calculations.

Microsoft Excel is an interpreted language, and therefore executes slowly. Moreover, the analysis systems are often run over an Apple Computer Localtalk network, making file access even slower. However, it has been found that automated analysis systems free the operator for other duties and still run fast enough to be quite acceptable.

At ANL-West we have found that the separation of the data acquisition software from the data analysis software has many advantages. Microsoft Excel's combination of spreadsheets, macros, and graphics allows the development of completely automated analysis systems for routine assays. The flexibility of the spreadsheet system allows easy processing of data from unusual assays and the visual programming interface makes processing system development fast and easy.

References

1. K. Debertin and R. G. Helmer, Gamma- and X-Ray Spectrometry with Semiconductor Detectors, Elsevier Science Publishers, 1988.

2. S. L. Meyer, Data Analysis for Scientists and Engineers, John Wiley & Sons, 1975.

3. W. H. Press et al, Numerical Recipes, Cambridge University Press, 1986.

4. J. L. Parker, "The Use of Calibration Standards and the Correction for Sample Self-Attenuation in Gamma-Ray Nondestructive Assay", LA-10045, Los Alamos National Laboratory, August 1984.