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AUTOMATED MULTISPECTRA ALPHA SPECTROMETER AND DATA REDUCTION SYSTEM

R. C. HOCHEL

SAVANNAH RIVER LABORATORY
AIKEN, SOUTH CAROLINA 29801

PREPARED FOR THE U.S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION UNDER CONTRACT AT(07-2)-1

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AUTOMATED MULTISPECTRA ALPHA SPECTROMETER AND DATA REDUCTION SYSTEM

by

R. C. Hochel

Approved by

E. L. Albenesius, Research Manager
Environmental Effects Division

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E. I. DU PONT DE NEMOURS AND COMPANY
SAVANNAH RIVER LABORATORY
AIKEN, SOUTH CAROLINA 29801

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ABSTRACT

A complete hardware and software package for the accumulation and rapid analysis of multiple alpha pulse height spectra has been developed. The system utilizes a 4096-channel analyzer capable of accepting up to sixteen inputs from solid-state surface barrier detectors via mixer-router modules. The analyzer is interfaced to a desk-top programmable calculator and thermal line printer. A chained software package including spectrum printout, peak analysis, plutonium-238 and plutonium-239 data reduction, and automatic energy calibration routines was written. With the chained program a complete printout, peak analysis, and plutonium data reduction of a 512-channel alpha spectrum are obtained in about three minutes with an accuracy within five percent of hand analyses.

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AUTOMATED MULTISPECTRA ALPHA SPECTROMETER AND DATA REDUCTION SYSTEM

INTRODUCTION

An expanded program for the measurement of plutonium in the environment of the Savannah River Plant has substantially increased the number of samples requiring alpha-spectrometric counting and analysis to determine plutonium isotopic compositions and contents. These analyses are particularly intended to provide information for assessing the environmental effects of the expected future growth of the nuclear industry in the southeastern United States. To cope with the present increased sample load and to provide capabilities for additional counting requirements of anticipated new programs, the existing counting systems were expanded and automated. A system, complete with hardware for processing up to sixteen alpha spectra and the software for performing energy calibration, data printout, spectrum peak analyses, and data reduction, is described in the following section.

INSTRUMENTATION AND DATA REDUCTION HARDWARE

The alpha pulse height analysis (APHA) system consists of a Model 8100 4096-channel analyzer and two Model 8220 four-input mixer-routers all supplied by Canberra Industries of Meriden, Connecticut. The mixer-routers allow eight 512-channel spectra to be simultaneously accumulated and stored in the analyzer memory. Additional mixer-routers may be interfaced to the analyzer allowing up to sixteen inputs.

Each of the eight inputs is obtained from a Model 485 amplifier from Ortec Incorporated of Oak Ridge, Tennessee, and is fed by an Ortec Model 125 preamplifier and a Series A surface barrier detector, 150 mm^2 area x 50-micron deep. The resolution for all the detectors is between 16 and 19 keV full-width, half-maximum (FWHM) for the 5.5-MeV alpha peak of americium-241. The detectors are biased by a selectable-polarity eight-pot adjustable power supply designed at the Savannah River Laboratory (SRL). The alpha detector vacuum chambers are of Nuclear Diodes (presently Edax International) design, fabricated at SRL.

ENERGY CALIBRATION PROGRAM

This program is essentially the same as the one supplied by APT with the DTU but rewritten to use the line printer. Using the linear equation relationship

$$E_i = aC_i + b \qquad (2)$$

where E_i is the energy of a peak with centroid C_i, a is the slope (keV or MeV/channel), and b is the channel-zero energy intercept; parameters a and b are calculated from the centroids of two known-energy peaks as determined from Equation 1. The operator has only to intensify the two peak regions and input their energies into the calculator. Upon completion of the calibration, the program stores the a and b parameters in two selected calculator registers for use in subsequent programs and prints their values on the line printer. This hard copy option is convenient for use where separate spectra are stored in different segments of memory, since a and b can be determined from a standard source count with individual detectors and listed as a permanent record of the energy calibration of each.

DATA PRINTOUT PROGRAM

Although automated spectral analyses are faster than hand calculations, hand calculations are generally more accurate and dependable. Even the most sophisticated reduction codes will occasionally encounter spectra which cannot be correctly analyzed. In such instances a hard copy of the data can be used. Fortunately the speed and flexibility of the calculator-line printer combination make hard copy output both fast and compact.

The operation of the data printout program is diagrammed in Figure 1. As mentioned previously, the first C_i, D_i read in from any region is always 0, T. Thus at the start of the program, the time is read in and then immediately loaded and printed.* The next read causes the first C_i, D_i of the intensified region to be transferred to the calculator. A check is then made for a negative C_i indicating the end of the region. Because the first transfer would not result in a negative C_i, calculator control passes on causing C_i and D_i to be loaded in the printer. The next read then occurs, and a check for a negative C_i is again made. If C_i is negative, control is transferred to cause a printout of the data already loaded, and the program stops; otherwise the D_i

---

* The printer, just as the DTU, is under calculator control. Alpha-numeric messages and/or data are first loaded by the calculator on a single 80-character line. The line can then be added to or printed.

The analyzer was interfaced to a HP (Hewlett-Packard Calculator Products Division of Loveland, Colorado) Model 9810A programmable desk-top calculator with a 2036-step capacity and 51 total data storage registers via an Applied Physical Technology (APT of Atlanta, Georgia) Data Transfer Unit (DTU). A HP 9866A thermal line printer interfaced to the 9810A calculator uses the calculator's alpha-numeric read-only memory option to provide hard copy of data and results.

Three general programs (energy calibrate, peak picker, and a single peak analysis routine) written by D. M. Walker of APT for use on the HP 9801 calculator are supplied with the DTU. These programs have been used to develop more specific user-tailored programs such as CETUS.[1] All of these programs use the algebraic method of determining a peak centroid (PC) and/or total integrated peak area from the expression

$$PC = \sum_i C_i D_i \Big/ \sum_i D_i \qquad (1)$$

where D_i is the data content in channel C_i of the spectrum. The programs referred to above, and most others written for the HP 9810A, use the internal printer option of this calculator and not the thermal line printer. Although rewriting such programs for use with the line printer is relatively simple, it requires more steps and will lengthen the program; however, the increased output speed, flexibility in data and message formating, legibility, and reduced paper usage make the revision worthwhile.

OPERATION

The DTU not only transfers data from the analyzer to the calculator, but also formats the data. Selection of data to be transferred from the analyzer memory is made by intensifying regions of interest with the analyzer's band intensification controls. Upon initiation of the transfer process, two numbers, C_i and D_i, are placed in the x and y working registers of the calculator, respectively. In every case, the first pair transferred is zero, T, where zero is channel number zero and T is the count time in seconds stored in that channel. Transfer then continues under calculator control sequentially as C_1, $D_1 \rightarrow C_2$, $D_2 \rightarrow \ldots$ C_n, D_n where C_n is the last channel in the intensified region. At the next requested transfer, the DTU will output a negative channel number signifying that the previous channel was the last of the region of interest. The negative channel can also be used to provide a software stop of the calculator program.

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---

* The printer, just as the DTU, is under calculator control. Alpha-numeric messages and/or data are first loaded by the calculator on a single 80-character line. The line can then be added to or printed.

of this pair is loaded. Following the transfer a counter is indexed by one and tested. If the counter is less than 9, the sequential transfer and testing continues with each D_i being loaded in the printer. When the index finally reaches 9, D_i is again loaded, but the line is also printed, and calculator control is transferred to (1) to start another line.

The result of the program is a printed page showing counting time on the first line followed by successive lines listing a channel number, the contents of that channel, and the contents of the next nine channels. A 512-channel spectrum can be printed out in about 40 seconds and uses only 9 inches of paper.

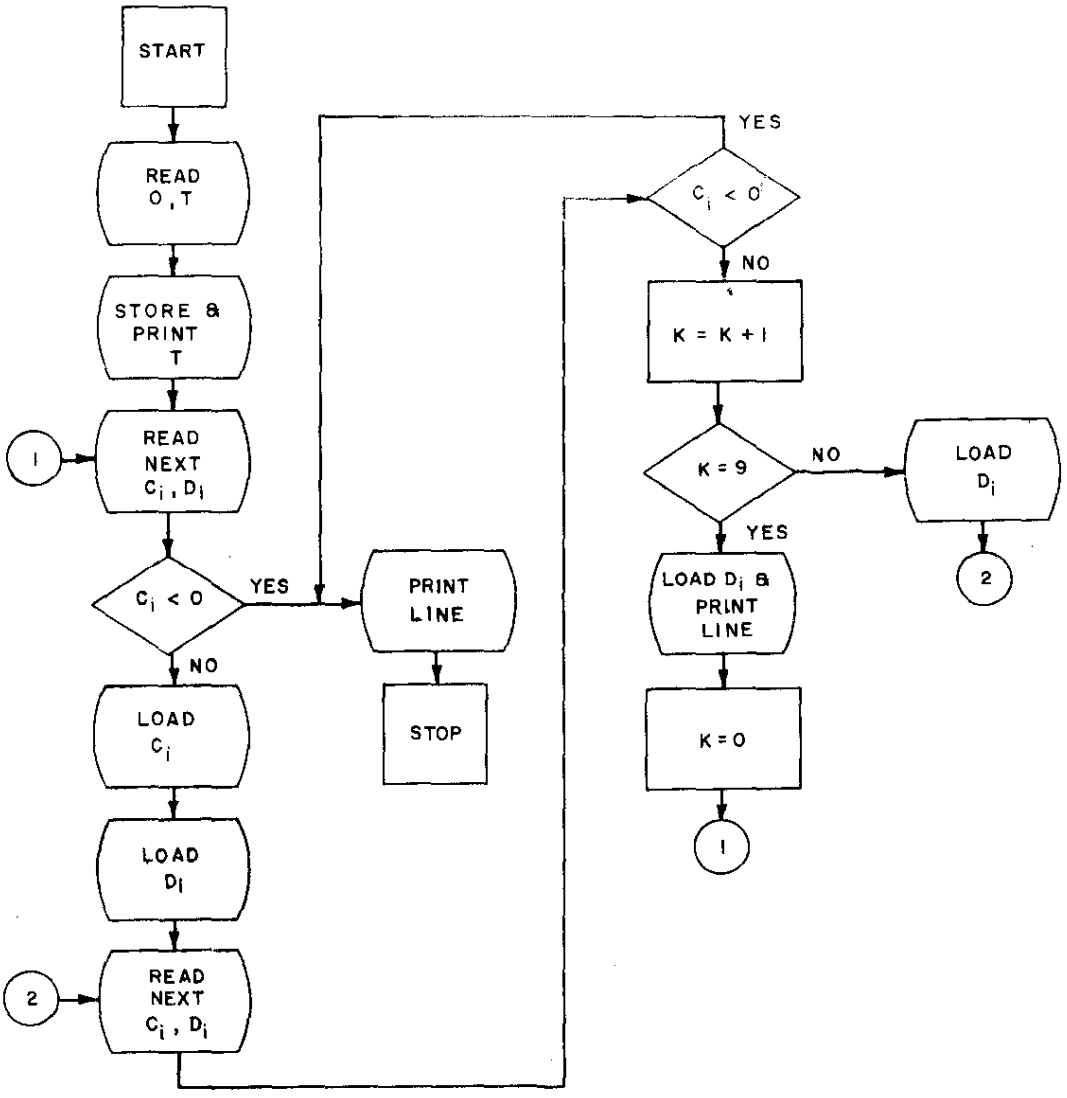


FIGURE 1. Data Printout Flow Chart

PEAK ANALYSIS PROGRAM

The peak analysis program consists of Parts A, B, and C (Figure 2). Part A selects the appropriate parameters a_i and b_i, previously determined from the energy calibration program or hand solutions of Equation 2, to be stored in the calculator for subsequent use in computing peak energies. The parameters are selected by inputing the octant number (1-8) of the spectrum to be analyzed which is then compared by a series of IF statements to determine the proper a_i, b_i pair to be stored in selected calculator data registers. This method of selection results in a considerable saving of register space (2 compared to 16) and simplifies retrieving these data later in the program since the same two registers are always referenced, regardless of the octant being analyzed.

Part B locates the peaks in the spectrum and calculates their energies, areas, and a one-sigma uncertainty for each peak area. This routine is similar to that used by APT and Wood and Palms[1] but was modified to handle the low-statistics spectra frequently encountered in alpha counting. Initially three parameters, a statistical sensitivity S, a maximum channel peak-width W, and a background addition factor BAF, are read from the program and stored, followed by a STOP, to allow changing of the parameters if desired. Next the counting time is read in, stored, and printed. The actual search for peaks is started by reading in the first six C_i, D_i's of the spectrum or intensified region. The first three D_i's are added to obtain a left-hand background continuum, which is used to test for the beginning of a peak in the 5th and 6th channels. The test is an IF statement of the form

$$1.5(D_{i-1} + D_i) > BKG \qquad (3)$$

where i-1 and i refer to the 5th and 6th channels, respectively, of the six-channel interval. BKG is determined from the expression

$$BKG = \sum_{i-5}^{i-3} D_i + \sqrt{S \sum_{i-5}^{i-3} D_i} + BAF \qquad (4)$$

Equations 3 and 4, aside from the BAF term, ask that the averaged contents of the i-1th and ith channels be greater than the averaged background continuum plus a factor $\sqrt{S/3}$ times its statistical fluctuation. The somewhat unusual forms of Equations 3 and 4 are used simply as a convenience in programming. Typically the value of S is set at 20 to 40, depending on background and desired sensitivity. The BAF term is added to avoid normal low-statistics background fluctuations from being interpreted as peaks since the first two terms of Equation 4 will frequently be zero in alpha spectra. A BAF of 5 or 6 yields

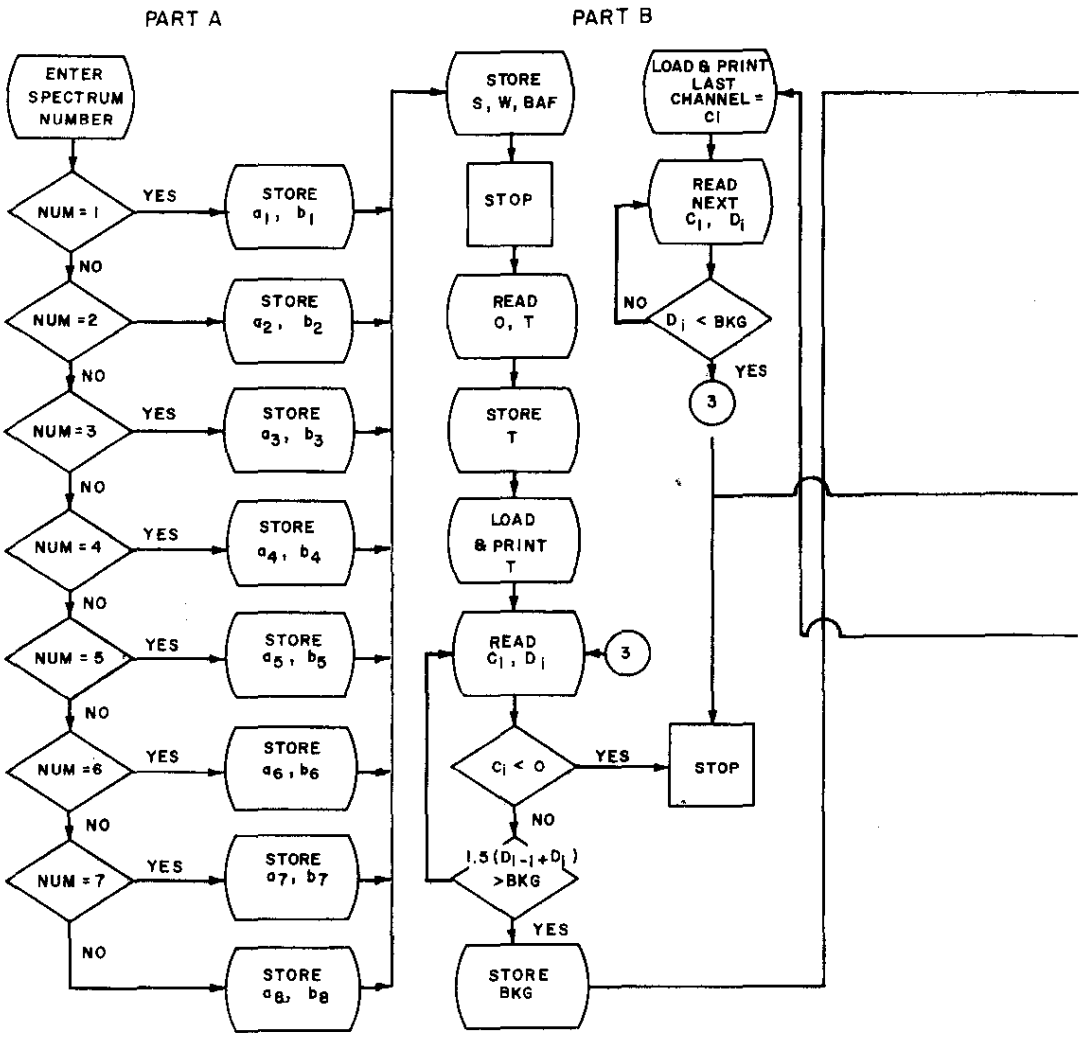


FIGURE 2. Peak Analysis Flow Chart

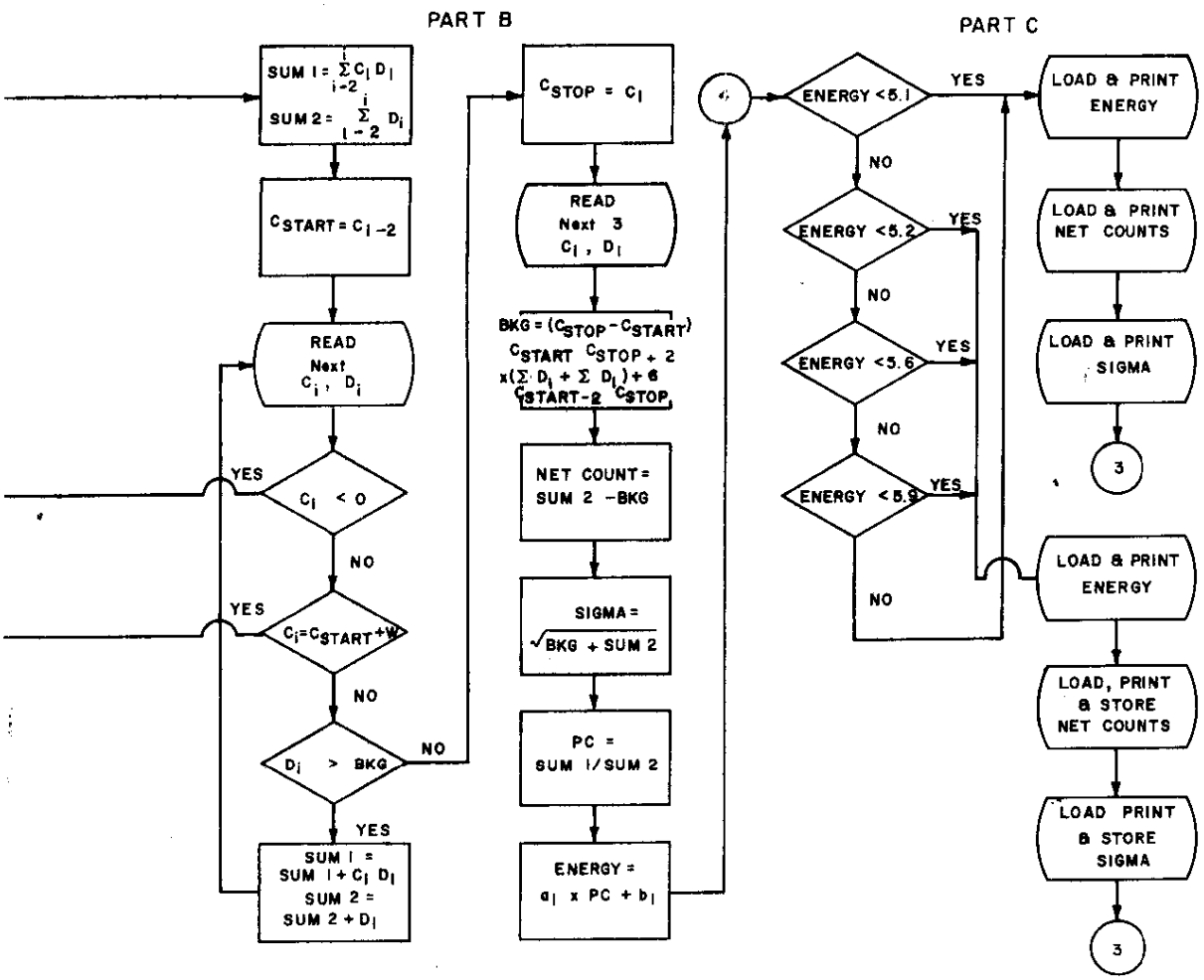


FIGURE 2. Cont'd.

the best results. Note also (Figure 2) that each C_i of the C_i, D_i pair read in is always tested for the end of a region to terminate the program.

If Equation 3 is false, the next C_i, D_i is read in, and the above process is repeated with the second through seventh channels. When Equation 3 is true, a peak is indicated, and the program proceeds to calculate SUM 1 and SUM 2 as shown in Figure 2 and also sets the starting peak channel (C_{START}) equal to C_{i-2}. Another C_i, D_i is read in and tested by

$$C_i = C_{START} + W \tag{5}$$

where W, the maximum peak width parameter, depends on peak resolution and intensity and is usually set at 20. If Equation 5 is true, the peak is too wide, and a message indicating so is printed out along with C_i, so the operator may later hand-integrate this peak and check for a doublet. The read-in process continues until a D_j is found which is less than BKG, whereupon the program returns to (3) to start a search for the next peak. If Equation 5 is false, D_i is tested to see if it is still greater than BKG. If it is, SUM 1 and SUM 2 are incremented, the next C_i, D_i is read in, and the loop continues until a D_i less than BKG is found. This signifies the end of the peak. C_i is noted as the last channel (C_{STOP}) in the peak, and then the next three C_i, D_i pairs are read in to form the right-hand continuum. A new BKG is next calculated using

$$BKG = (C_{STOP} - C_{START}) \left(\sum_{C_{START-2}}^{C_{START}} D_i + \sum_{C_{STOP}}^{C_{STOP+2}} D_i \right) / 6 \tag{6}$$

The values for net counts in the peak, sigma, peak centroid, and peak energy are determined from the appropriate expressions in Figure 2.

Part C of the peak analysis program facilitates use of a final program using peak characteristics as determined by part B to calculate desired isotopic plutonium concentrations. While this last program is tailored specifically for the type of plutonium analyses being performed in this laboratory, it can easily be modified to treat data from other alpha emitters.

The routine (Part C) checks the energies of each of the peaks as they are detected against the 5.15-MeV and 5.50-MeV alpha peaks of ^{239}Pu and ^{238}Pu, respectively, and also the 5.75-MeV peak of ^{236}Pu, which is a spike isotope added to determine the plutonium chemical yield through separation and source preparation. In reality all of these plutonium peaks are doublets, but the detector resolution and source thickness usually do not

allow complete separation of the peaks, and the program will see them as singlets.

The routine labeled (4) in Figure 2 shows the logic involved. The energy of the detected peak is first checked to determine whether it is less than 5.1 MeV. If it is less, the energy, net counts, and sigma are printed out on the line printer, and control returns to (3) to continue the search. If energy is greater than 5.1 MeV, a check is made against 5.2 MeV. If the energy is less than 5.2 MeV, the peak is assumed to be ^{239}Pu, and the net counts and sigma are stored and then printed along with the energy. This process, by continuation, stores and prints peak characteristics for ^{238}Pu and ^{236}Pu. Any peaks greater than 5.9 MeV are just printed. Thus at the end of the peak analysis program, a listing of the energies, net counts, and sigmas for all the peaks in the spectrum is obtained on the line printer, and the net counts and sigmas for the plutonium peaks are also stored in six fixed data registers of the calculator and can be recalled in any subsequent program. About 1½ minutes are required to analyze a 512-channel spectrum, and results generally agree within five percent of hand calculations. Occasionally, severe low-energy tailing of peaks will cause erroneous peak integrations, but such cases are usually obvious, and hand calculations can be made. Because the program does not smooth the data, results are usually closer to hand analyses, especially for low-statistics spectra, than more-sophisticated routines. However, the program frequently detects psuedo-peaks, thus requiring more careful scrutiny of results.

DATA REDUCTION PROGRAM

The printout and peak analysis programs are standalone routines which can be used independently. However by chaining them together with a final routine (Figure 3), the data are converted to isotopic plutonium activities per gram of sample, total plutonium, and percent ^{238}Pu to make a complete alpha spectrum data reduction program.

At the start of the program, a number of parameters needed to perform the calculations are requested. The first three are weights used to find activities on a per gram basis. Because samples are dried and ashed, and only a portion of the ashed sample may be counted, the actual weight needed for the calculation is

$$Wt = \frac{\text{Dry Wt} \times \text{Sample Wt}}{\text{Ash Wt}}$$

Two other parameters requested and stored are the pCi activity of the ^{236}Pu spike added to the sample and a correction factor (previously obtained from standards for each detector) which converts counts/min to pCi. As each of these parameters is requested, the entered value is also written out as a record. The calculation proceeds as shown in Figure 3. At the end, control is returned to the printout routine so that the program is ready to start on another spectrum.

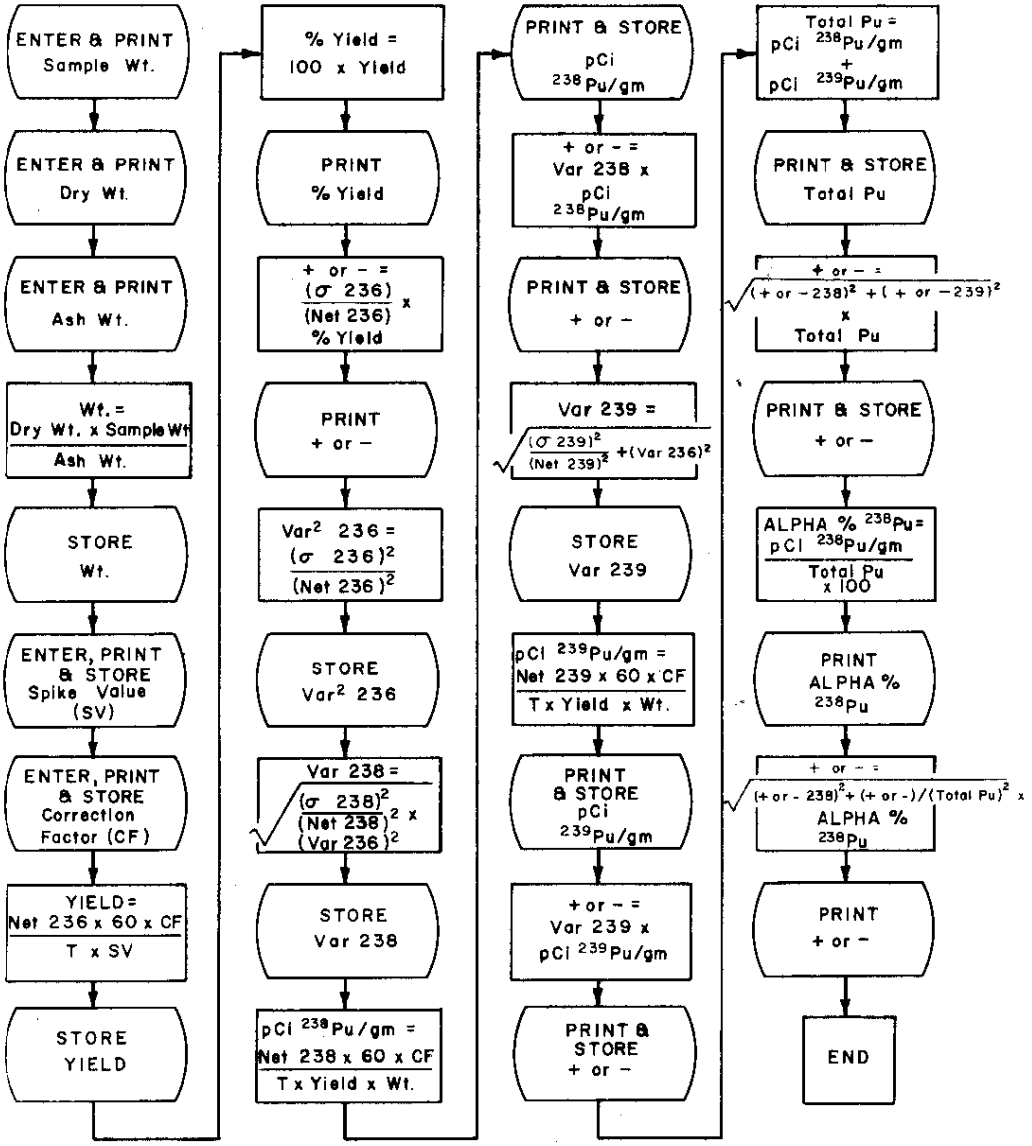
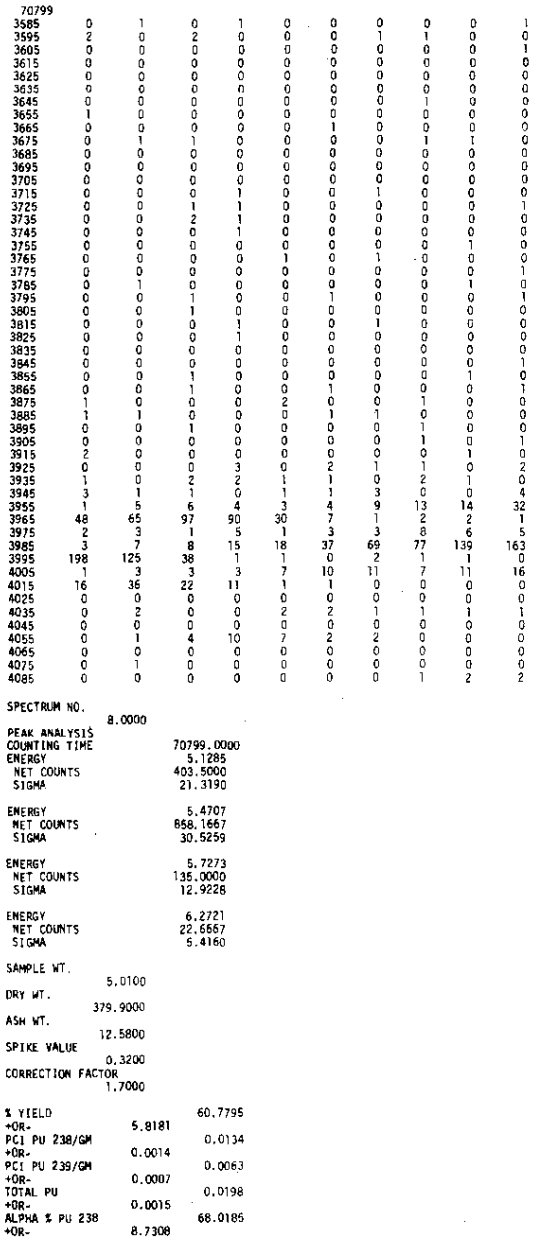


FIGURE 3. Data Reduction Flow Chart

A complete printout of the chained alpha spectrum data reduction program for a typical 512 channel plutonium count is shown in Figure 4 and was obtained in about three minutes, including requested data entry from the operator.



```
 70799
3585    0    1    0    1    0    0    0    0    0    1
3595    2    0    2    0    0    0    1    1    0    0
3605    0    0    0    0    0    0    0    0    0    1
3615    0    0    0    0    0    0    0    0    0    0
3625    0    0    0    0    0    0    0    0    0    0
3635    0    0    0    0    0    0    0    0    0    0
3645    0    0    0    0    0    0    0    1    0    0
3655    1    0    0    0    0    0    0    0    0    0
3665    0    0    0    0    0    1    0    0    0    0
3675    0    1    1    0    0    0    0    1    1    0
3685    0    0    0    0    0    0    0    0    0    0
3695    0    0    0    0    0    0    0    0    0    0
3705    0    0    0    0    0    0    0    0    0    0
3715    0    0    0    1    0    0    1    0    0    0
3725    0    0    1    1    0    0    0    0    0    1
3735    0    0    2    1    0    0    0    0    0    0
3745    0    0    0    1    0    0    0    0    0    0
3755    0    0    0    0    0    0    0    0    1    0
3765    0    0    0    0    1    0    1    0    0    0
3775    0    0    0    0    0    0    0    0    0    1
3785    0    1    0    0    0    0    0    0    1    0
3795    0    0    1    0    0    1    0    0    0    1
3805    0    0    1    0    0    0    0    0    0    0
3815    0    0    0    1    0    0    1    0    0    0
3825    0    0    0    1    0    0    0    0    0    0
3835    0    0    0    0    0    0    0    0    0    0
3845    0    0    0    0    0    0    0    0    0    1
3855    0    0    1    0    0    0    0    0    1    0
3865    0    0    1    0    0    1    0    0    0    1
3875    1    0    0    0    2    0    0    1    0    0
3885    1    1    0    0    0    1    1    0    0    0
3895    0    0    1    0    0    0    0    1    0    0
3905    0    0    0    0    0    0    0    1    0    1
3915    2    0    0    0    0    0    0    0    1    0
3925    0    0    0    3    0    2    1    1    0    2
3935    1    0    2    2    1    1    0    2    1    0
3945    3    1    1    0    1    1    3    0    0    4
3955    1    5    6    4    3    4    9   13   14   32
3965   48   65   97   90   30    7    1    2    2    1
3975    2    3    1    5    1    3    3    8    6    5
3985    3    7    8   15   18   37   69   77  139  163
3995  198  125   38    1    1    0    2    1    1    0
4005    1    3    3    3    7   10   11    7   11   16
4015   16   36   22   11    1    1    0    0    0    0
4025    0    0    0    0    0    0    0    0    0    0
4035    0    2    0    0    2    2    1    1    1    1
4045    0    0    0    0    0    0    0    0    0    0
4055    0    1    4   10    7    2    2    0    0    0
4065    0    0    0    0    0    0    0    0    0    0
4075    0    1    0    0    0    0    0    0    0    0
4085    0    0    0    0    0    0    0    1    2    2

SPECTRUM NO.
          8.0000
PEAK ANALYSIS
COUNTING TIME              70799.0000
ENERGY                         5.1285
 NET COUNTS                  403.5000
 SIGMA                        21.3190

ENERGY                         5.4707
 NET COUNTS                  858.1667
 SIGMA                        30.5259

ENERGY                         5.7273
 NET COUNTS                  135.0000
 SIGMA                        12.9228

ENERGY                         6.2721
 NET COUNTS                   22.6667
 SIGMA                         5.4160

SAMPLE WT.
          5.0100
DRY WT.
        379.9000
ASH WT.
         12.5800
SPIKE VALUE
          0.3200
CORRECTION FACTOR
          1.7000

% YIELD                       60.7795
+OR-          5.8181
PCI PU 238/GM                  0.0134
+OR-          0.0014
PCI PU 239/GM                  0.0063
+OR-          0.0007
TOTAL PU                       0.0198
+OR-          0.0015
ALPHA % PU 238                68.0185
+OR-          8.7308
```

FIGURE 4. Typical Program Printout

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Copy No.

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