Sample Preparation and Gamma-ray Spectrometer Operation for Determining Natural Radioelement Contents in Rocks at the U.S. Geological Survey in Denver, Colorado

by

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Open-File Report 81-1308

1981

INTRODUCTION

Gamma-ray spectrometry is used by the Radioelement Distribution Project of the USGS for the quantitative analysis of the naturally occurring radioelements (uranium, thorium, and potassium) in rocks and soils. Potassium is determined by measuring ^{40}K which is considered to be a constant percentage of the total potassium. Thorium is indirectly determined by measuring its daughters ^{212}Pb, ^{212}Bi, and ^{208}Tl. The half-lives of the ^{232}Th daughters are short and the thorium decay series is almost always in equilibrium. Uranium concentration is determined by measuring short-lived daughters (^{214}Pb and ^{214}Bi) of ^{226}Ra; the results are reported as radium-equivalent uranium (RaeU). RaeU is the amount of uranium required to support the measured amount of radium in secular equilibrium. Because of disequilibrium between ^{238}U and ^{226}Ra in the decay series, this value may not necessarily be the same as the actual amount of uranium present, particularly in soils or near-surface rocks.

These data are then used for many types of basic and applied research programs. Some examples are: 1) Studies of the processes causing uranium ore bodies to be formed; 2) exploration for and evaluation of uranium or thorium resources; and 3) research into analytical techniques to improve precision and detection limits of analyses for the naturally occurring radioelements.

This manual is written as an explanation of procedures used by the Radioelement Distribution Project to determine concentrations of the radioelements in rocks and soils. It is intended for new operators of the equipment in the project. The mention of trade names does not imply an endorsement of the products by the U.S. Geological Survey. The manual covers methods of sample preparation, analyzer operation, and computer operation to obtain the most accurate analyses and to maintain the best precision possible for radioelement analyses. Although the analyses are interpreted by computer, the calibration methods and operational procedures are similar to Bunker and Bush (1966, 1967).

THEORY

When a photon is absorbed by a scintillator such as thallium-activated sodium iodide, NaI(Tl), a pulse of light is emitted by the scintillator. The intensity of the light is proportional to the energy of the photon. A photomultiplier tube optically bonded to the NaI(Tl) crystal sees the pulse of light and converts it to an electrical signal. The voltage of the signal is proportional to the intensity of the light; therefore it is proportional to the energy of the photon. That electrical pulse is amplified, processed by an analog-to-digital converter (ADC), and input to a multichannel pulse height analyzer. The analyzer sorts the pulses by voltage, moving a pulse to a channel proportional to the voltage of the pulse. Each time a pulse is assigned to a channel, the number of counts in that channel is incremented by one. Therefore, each channel of the analyzer is proportional to the voltage of the pulses included in that channel and to the energy of the photon producing those pulses. The number of counts accumulated in that channel per unit of time is proportional to the number of disintegrations producing gamma-rays at that energy during that time, and to the quantity of that isotope present in the sample.

A gamma-ray emitting radioisotope produces one or more gamma-rays of specific and usually distinctive energies; however, several isotopes may emit gamma-rays at a given energy. In a large number of atoms of a radioisotope, a certain fraction will always emit gamma-rays per unit of time. Knowing the energies emittted by an isotope may identify the isotope, and knowing the rate the gamma-rays are emitted enables one to calculate the quantity of the isotope present in the sample. However, to accurately calculate the concentration of isotope in the sample, measured values must be compensated for absorption of gamma-rays within the sample and for the geometry of the sample relative to the detector.

HARDWARE

The NaI(Tl) counting system (fig. 1) consists of sixteen 12.7 cm (5 in) diameter by 10.2 cm (4 in) thick NaI(Tl) crystals integrally connected to 12.7 cm diameter photomultiplier (PM) tubes. Each detector is located within a lead shield with 10.2 cm thick walls and interior dimensions of 40.6 cm (16 in) x 40.6 cm x 71.1 cm (28 in). Preamplifiers are plugged on the other end of the PM tubes. Each preamplifier is connected by a coaxial cable to a linear amplifier mounted in Nuclear Instrument Module (NIM) bins. The output signals from four amplifiers are all connected to a mixer-router where the signals are multiplexed and output on one cable. The outputs from two mixer-routers are connected so that one cable, carrying the signals from eight detectors, is input to a Canberra Model 8060 ADC then to a Canberra Model 8100 4096-channel pulse-height analyzer (PHA). Each detector is assigned 512 channels of analyzer memory. The external ADC converts the electrical signals to digital values, sorts the pulses by detector, and stores them by energy in the analyzer memory. Two pulse-height analyzers accommodate all sixteen detectors. Potentiometers on each amplifier vary the gain of each detector so that each 512 channel segment represents an energy range from 0.1 to 1.9 million electron volts (mev).

Each PHA is connected to the same Texas Instruments (TI) Model 733 electronic data terminal through a switch. A Canberra Model 1482 Data Input device is switched between both multichannel analyzers so that four 6-digit

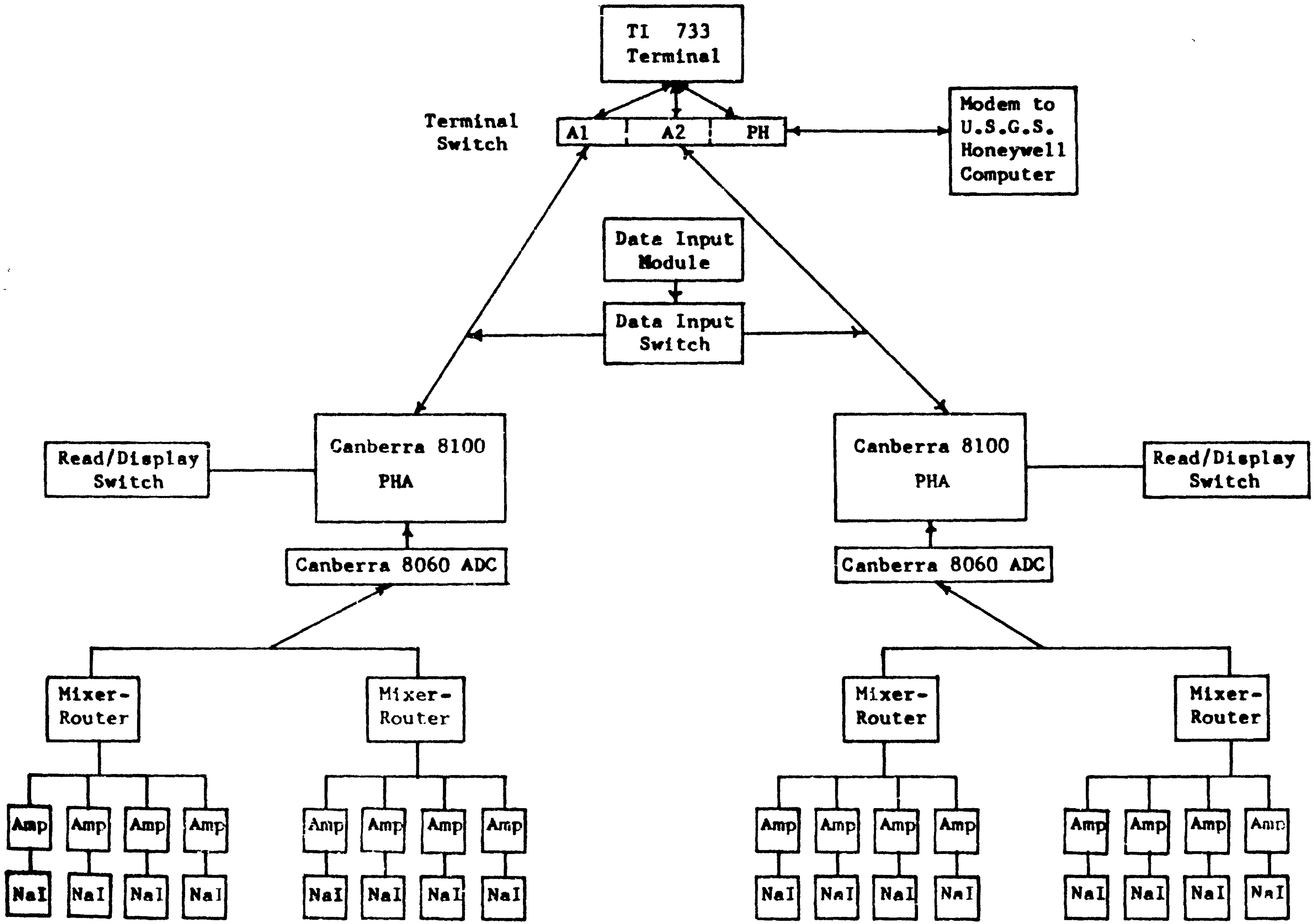

Figure 1. A block diagram of the scintillation counting equipment of the Natural Radioelement Distribution project of the U.S.G.S.

identification numbers are sent to the data terminal ahead of the spectral data. Data are transferred from the analyzer to the data terminal at 1200 baud (approximately 120 characters per second) through an EIA RS-232 Serial Data Interface, where they are stored on magnetic cassette tapes.

Data on the cassettes may be printed at the data terminal or transferred over telephone lines to the USGS Honeywell computer and stored on a mass storage disk. Spectra on the disk may then be edited and operated on by a computer program to interpret the digital data and calculate the concentrations of radioelements present in the sample.

SOFTWARE

The Alpha-M computer program used to interpret digital data from the analyzer is a modification of one written by Schonfeld (1966). The program is a linear least squares fitting program which compares a spectrum with a library of spectra stored in the computer. The library consists of 1) a spectrum of uranium isotopes present in natural abundances in equilibrium with their short-lived daughters, and with virtually none of their long-lived daughters, i.e. ^{226}Ra, (fig. 2), 2) radium with daughters in equilibrium, 3) thorium in equilibrium with daughters, 4) potassium, and 5) background. Output from the computer program lists the concentrations of RaeU, in parts per million (ppm), thorium in ppm, and percent potassium. The program also prints the standard error of each measurement and the results of a chi-square test to determine the "goodness of fit" of the library standards to the unknown spectrum. Figure 3 is a spectrum of a rock sample showing typical thorium, uranium, and potassium concentrations.

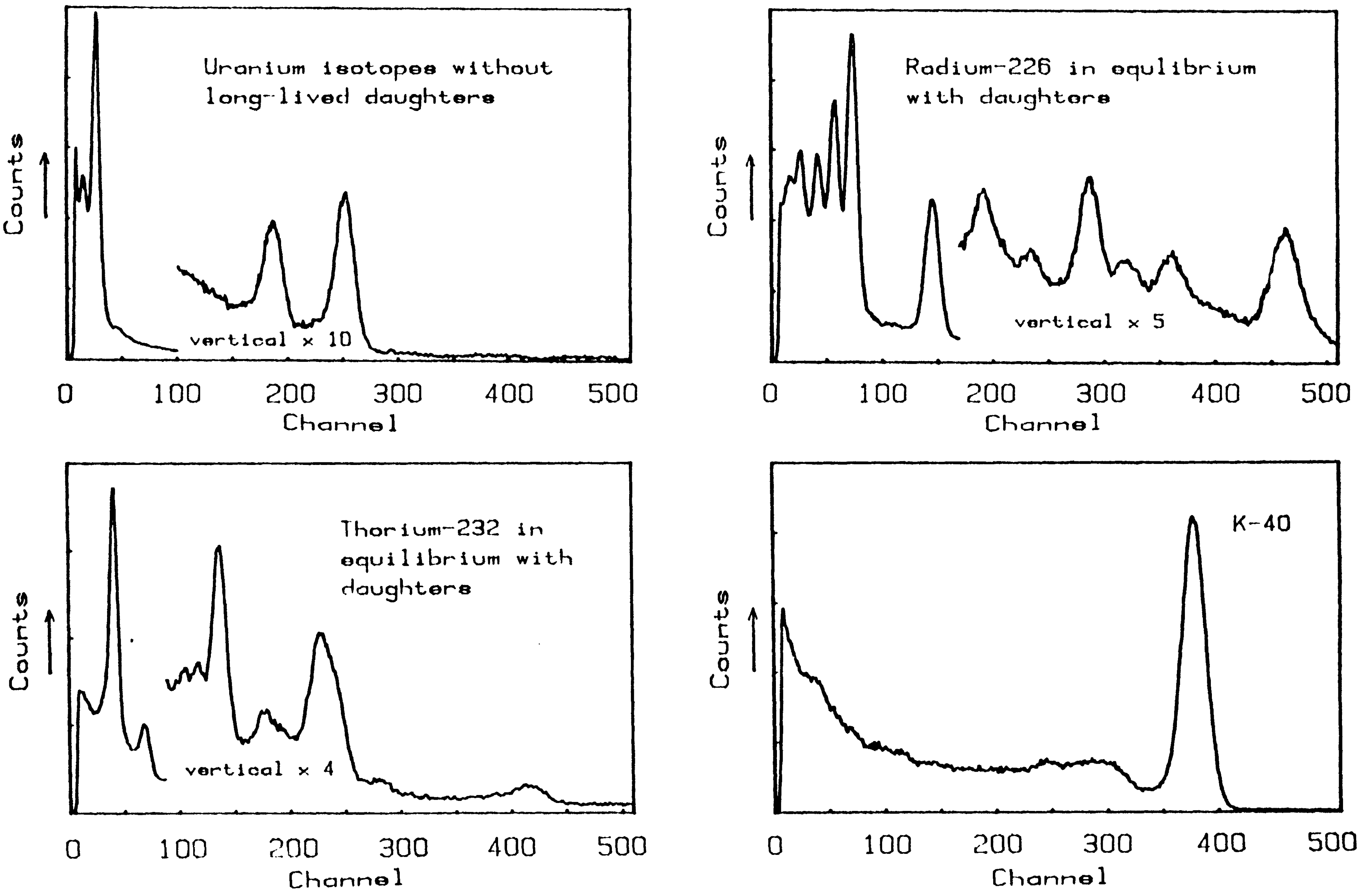

Figure 2. Spectra of library standards used by the Alpha-M computer program to quantitatively interpret gamma-ray radioelement spectra of samples with unknown concentrations.

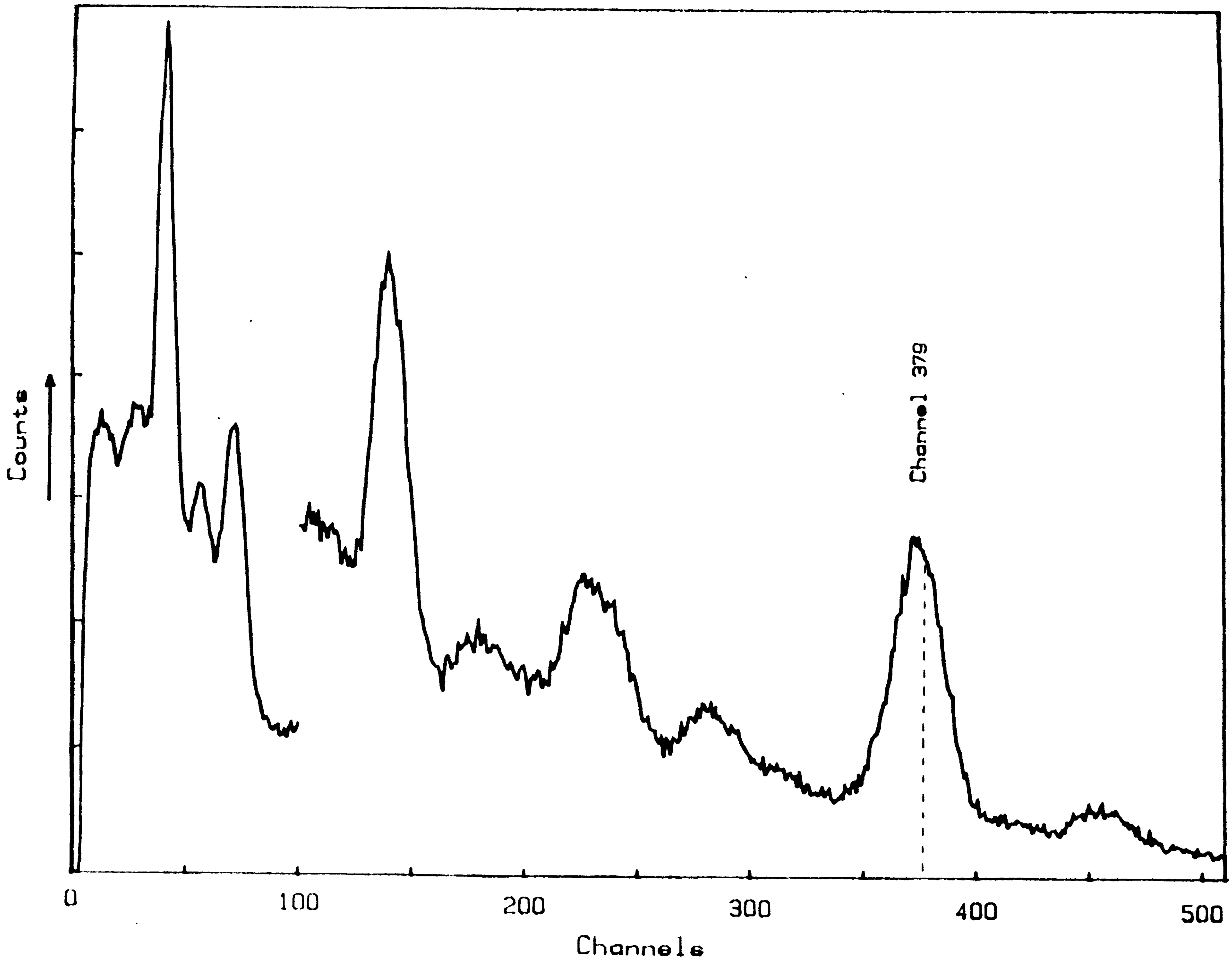

Figure 3. A spectrum of gamma-rays from a rock with typical radioelement ratios.

SAMPLE PREPARATION

Samples should be pulverized to minus 30 mesh (less than about 0.5 mm) for high quality analytical results; however, larger diameter material such as cuttings from drill holes are sometimes used without further preparation when lower quality results are acceptable.

After grinding, 600 g of most samples are weighed into 15.2 cm (6 in) diameter 2.5 cm (1 in) thick plastic containers. Exceptionally low density samples must be weighed into containers that are 3.8 cm (1.5 in) deep to contain the entire 600 g. If the available sample is less than 600 g, but more than 400 g, the entire sample is encapsulated. The containers are then sealed around the edges of the lids with plastic electrical tape to prevent loss of radon (^{222}Rn) and thoron (^{220}Rn).

If less than 400 g of sample is available or if the sample is very radioactive, the sample is weighed into a 4-dram glass vial 5.1 cm (2 in) high by 2.2 cm (0.9 in) diameter. Enough sample is placed into the vial so that after compacting the sample, the height of the sample is about 4.6 cm (1.8 in).

After encapsulation, the bulk density of the sample must be determined. The samples are placed on edge and a vibrator is rubbed a few times across each side of the container to level the surface (fig. 4), to remove air pockets from the sample, and to compact the sample to approximately the same density it will be when the container is laid flat and the upper surface leveled. The most important aspect of vibrating the sample is to be consistent and treat each sample alike. A template (fig. 4) has been prepared for both the 1 in and 1-1/2 in containers. The template was prepared by placing measured volumes of water into an empty container and measuring the height of the water in the container. Bulk density is then calculated by

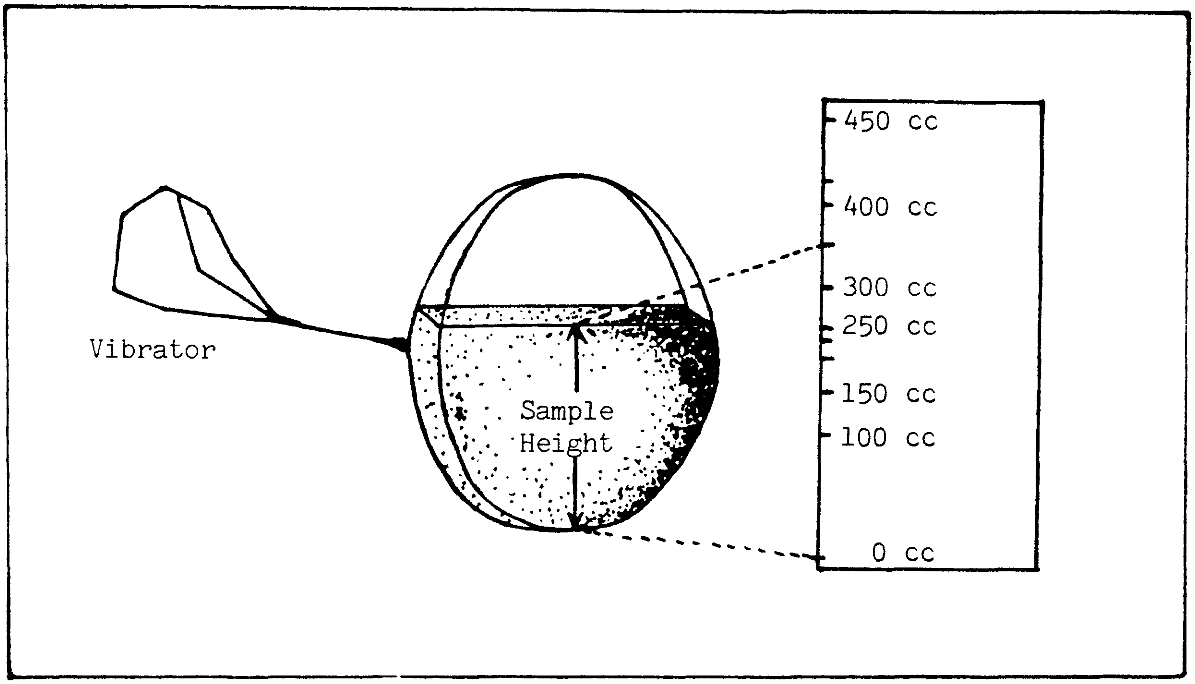

Figure 4. Position of the sample container while being vibrated to compact the sample and to compare sample height to the volume template to determine the volume occupied by the known weight of samples.

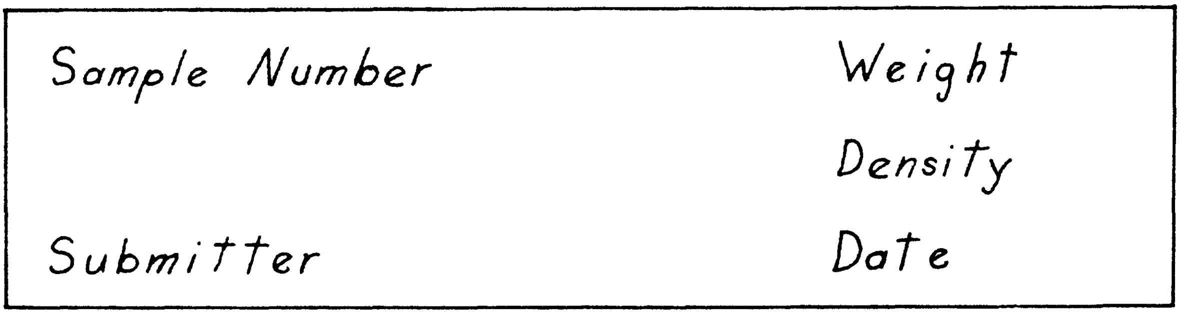

Figure 5. Location of items to be listed on sample labels.

dividing the weight of the sample by the volume of water representing the height of the sample on the template.

A constant that is proportional to the density of the sample in the vials is calculated by measuring the weight of sample in the vial and dividing by the height of the sample. This constant is used as the density value in sample corrections.

A label (fig. 5) is then made by writing legibly with indelible ink, the sample number, weight of sample, density of sample, submitter, and date of encapsulation on a piece of masking tape and placing it on the lid of the sample container. This procedure insures maintaining the traceability of the sample from preparation through data reporting.

After all samples in a group from one submitter have been encapsulated and their densities measured, the samples are ordered by field or laboratory number, and their weights and densities, with all other available information, are tabulated on a Radioelement Analysis Request form (fig. 6).

The samples are then set aside for about three weeks before counting to allow the short-lived radioisotopes to reach secular equilibrium with their long-lived parents.

OPERATING PROCEDURES

The optimum count time for a sample is the amount of time required for the maximum number of counts in any channel of the PHA to reach about 6000 counts. This occurs when the channel is about two-thirds of full scale when the full-scale vertical range is set to 10^4 (fig. 7). When the analyzer is accumulating, the elapsed live-time can be read at the top-center of the cathode-ray tube (CRT). To estimate the optimum count time for a sample, a spectrum of the sample is accumulated for a short time and the counts

RADIOELEMENT ANALYSIS REQUEST

Project Area ____________________

Submitted by: ____________________ Date: ____________________ Collected by: ____________________

Field and Lab. No.	Coordinates	State and County	Formation	Sample Description, Geologic Setting, Age	Wt.(g)	Dens.

Figure 6. Radioelement Analysis Request form.

accumulated per second are extrapolated until the optimum number of counts would be accumulated. The vertical range switch should be set to 10^2 so that full-scale is 100 counts. Accumulate a spectrum until the center channel of the largest peak reaches full scale. The optimum count time is about 60 times the number of seconds that were required to accumulate the 100 counts.

To accumulate spectra for eight detectors in an analyzer: 1) Samples should be shaken to thoroughly mix grain sizes; then the upper surface of the sample leveled by gently dropping flat on a table top several times. 2) The samples are placed on the face of the detectors, centering the detectors in the ring on the bottom of the sample containers. 3) The preset live time thumbwheel switches on the analyzer are set to the desired counting time. The left switch is labeled N and the right switch is labled M. Count time is determined by $N \times 10^M$ seconds . 4) The Memory Control switch is set to 1/1 and Memory Subgroup to Off. 5) The READ/DISPLAY switch on the center NIM bin is set to read, then the COLLECT button is pressed until it lights up.

When the analyzer has counted for the preset time, the light in the COLLECT button will go out; or, if sufficient counts have been accumulated, the COLLECT button should be pressed to stop the data collection.

After a spectrum has been accumulated, it should always be determined if there has been a gain change. There is generally a large K-40 peak that should be centered on channel 379. If the peak is not in the proper location, the fine-gain potentiometer on the linear amplifier should be adjusted so that the next spectrum accumulated will be located properly. Turning the potentiometer clockwise moves the peak to the right (increasing energy) and turning it counter clockwise moves it to the left. A spectrum may be used if the peak is within about 5 channels of the proper locations since the Alpha-M computer program will shift the spectrum to the proper gain before interpreting the data.

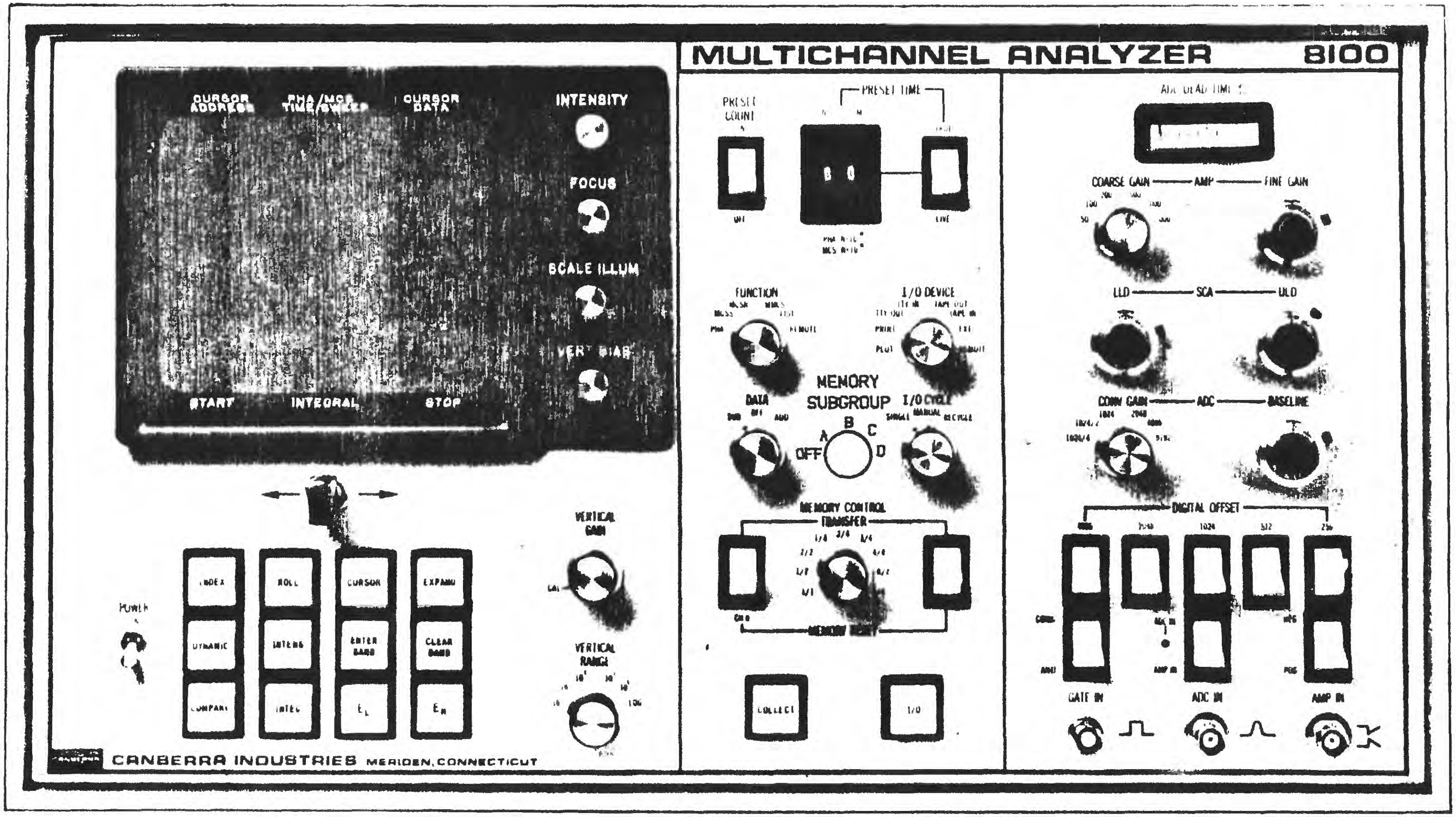

Figure 7. Front panel of Canberra Model 8100 multichannel pulse-height analyzer.

To display, readin or readout (I/O), or erase a 512 channel memory group from a single detector, the READ/DISPLAY switch in the center NIM bin must be set to READ. The analyzer switches require the following settings:

DETECTOR NUMBER	MEMORY SUBGROUP	MEMORY CONTROL
1	A	1/2
2	A	2/2
3	B	1/2
4	B	2/2
5	C	1/2
6	C	2/2
7	D	1/2
8	D	2/2

It is important to remember that I/O or erase switches only apply to the memory subgroups that appear in the CRT on the front of the analyzer.

Before reading out a spectrum to tape, first complete an entry in the sample logbook (fig. 8). The entry assigns a tape number to each sample and lists the count time, sample weight, and sample density. Tape numbers are ordered sequentially from 0000 to 9999. These data are typed at the end of the digital data and may be used as a check to insure that the proper values are dialed on the Data Input Module thumbwheel switches (fig. 9) whose values precede the digital spectral data.

To read out a spectrum to a cassette tape in the data terminal: 1) The terminal switch (fig. 9) must be turned toward the multichannel analyzer being read out. 2) The Data Input switch is turned toward the analyzer being read out. 3) The RECORD device function switch on the terminal is set to the LINE position. 4) The PLAYBACK/RECORD switch is set to RECORD for the cassette on the side of the terminal adjacent to the analyzer to be read out. The RECORD

Sample Number	Tape Number	Counting Time(sec)	Weight (Grams)	Density (G/CC)	Crystal	Date Analyzed	Energy Range	Remarks
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	
					Xtal		Mev	

Figure 8. A page from the logbook that is to be filled in when a sample spectrum is recorded on tape.

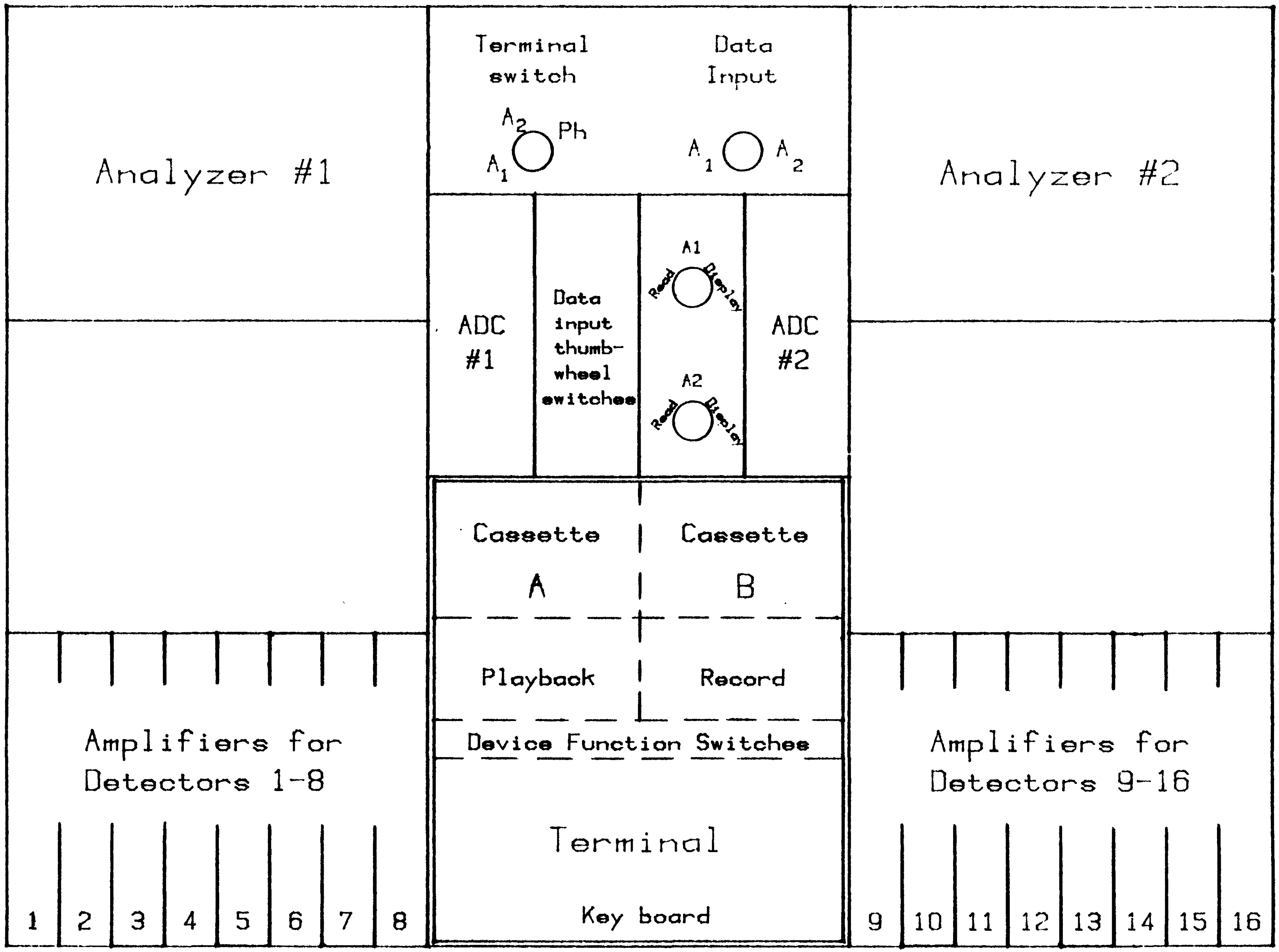

Figure 9. Diagram of the position of electronic equipment in the instrument racks.

CONTROL switch is set to ON. 5) The Memory Subgroup and Memory Control switches on the analyzer must then be set to the spectrum to be read out. 6) The thumbwheel switches on the Data Input module are set to put four tagwords on tape at the beginning of the spectrum. The top tagword on the Data Input Module is the detector number (two digits) followed by the four digit tape number that was assigned when the sample was identified in the logbook. The second tagword is the counting time of the spectrum in seconds. The third tagword is the sample weight multiplied by 100. The fourth tagword is the sample bulk density multiplied by 10^5. This means that an imaginary decimal point is located before the two least significant digits of the weight and after the most significant digit of the density thumb wheel switches. 7) The I/O button on the PHA should be pressed until it lights up. The lights in the RECORD CONTROL section of the terminal will flash quickly until the sample readout is finished. When the sample data are stored on the cassette, all lights will light up and the spectrum will reappear on the CRT. At that time, the RECORD CONTROL switch is set to OFF. 8) The RECORD device function switch is then set to local and the RECORD CONTROL switch is set to ON. The sample name and the digital data from the sample logbook should then be typed at the terminal. All data should be typed in the order that they appear on the line in the logbook. The RECORD CONTROL switch is turned to OFF. 9) To read out additional spectra, start with 3) above.

COMPUTER OPERATION

After the spectral data have been recorded on cassette, they must be transmitted to the USGS Honeywell computer to be interpreted.

To link the terminal to the computer: 1) The Terminal switch (fig. 9) is turned to the PH (telephone) position. 2) The terminal device function switches, KEYBOARD, PLAYBACK, and PRINTER must be set to the LINE position. 3) The TALK button on the modem atop the instrument rack is depressed and the 300 baud telephone number written on the modem should be dialed. When the high-pitched carrier signal is heard, the red button on the modem should be depressed. 4) The line feed (↑) key or the carriage return key (↓) on the terminal is pressed and the computer will respond by typing the number of people using the computer. "CBush↓" is typed and the computer will respond "PASSWORD". After the password has been typed, the computer will log the user on the system. It will list the terminal commands previously entered into the computer to control the operation of the Radioelement Distribution Project terminal.

To transmit the digital data from the cassette to the computer, the PLAYBACK/RECORD switch on the terminal must be set to PLAYBACK toward the cassette to be transmitted. "stty -modes ^lfecho,^crecho↓" is typed to prevent the computer from inserting an extra carriage return following each line. The QEDX editor in the computer is entered by typing "qx↓↑". The editor is placed in the input mode by typing "i↓↑". The data may be transmitted to the computer by pressing the CONT(inuous) START switch in the PLAYBACK CONTROL section of the terminal. The cassette will then advance, transmit the data to the computer, and print the data at the terminal. To suspend the printing of data, the PRINTER device function switch is moved to OFF. When the cassette has transmitted all the data, the end-of-tape light will be lit. To indicate to the editor that the transmission is finished, type "\f↓↑" (backslash) must be typed. Then the data from the editor's buffer is written into the CBush computer area after typing "w filename↓↑". Filename is generally the tape number of the sample at the beginning of the cassette. The editor is exited when "q↓↑" (quit) is typed. The command "stty -modes lfecho,crecho↓↑" causes the computer to resume normal terminal operations. Figure 10a is a listing of a sample spectrum as entered into the editor.

After the spectral data has been stored in the computer, it must be edited to put it in a format that can be read by the Alpha-M computer program. The easiest way to do this is by using another editor named TECO. Enter the TECO editor by typing "te filename↓". After each command has been completed the computer will respond "Z". The following commands are necessary to edit the file. The command "0j" sets the editor at the beginning of the file, "k" (kill) deletes a line, "d" (delete) deletes a character, "s/text/" searches for the specified text, "<ms/text//new text/>" replaces text with new text wherever the specified text is found in the file, "l" (line) advances a line, and "t"(type) prints a line. Commands may be strung together and are executed by typing "$↓".

To edit a spectral file, the following steps are necessary: 1) Enter the file with the editor by typing "te filename↓". 2) Because the first line is always blank, it should be deleted by typing "0jk0lt$↓", and the computer will respond by printing the first line of data. 3) Delete any blank lines in the file by searching for multiple carriage returns or carriage returns followed by line feeds, and replace them by single carriage returns using the command "0j<ms/↓↓//↓/>0j<ms/↓↑//↓/>$↓". 4) The spaces between channels are deleted by typing "0j<ms/ 0//0/$;>$↓0j<ms/ 0// 0/$;>$↓". 5) Delete the last channel of each spectrum by typing "0j<s/./018dl>$↓". 6) Print each spectral identification line by typing "0j621t<631t>$↓". This prints the line following each spectrum. When printing is finished, the print should be checked for typing errors and if any are found corrected using the ms command. 7) The editor is exited by typing "mweq$↓". Figure 10b is a listing of the sample spectrum after it has been edited.

```
024413
009000
060000
175000
012817
0001
017924 020686 020447 020398 016922 012428 009019 007326 006493 006303
006127 006605 006676 006426 006421 006345 006100 005919 005772 005787
005735 005904 005821 005920 006234 006361 006522 006837 007505 008690
010214 011418 011731 010657 008940 007381 005722 004951 004465 004280
004205 004165 004231 004082 003838 003879 003821 003925 004276 004583
004612 004571 004309 003981 003653 003194 002877 002640 002517 002267
002266 002232 002226 002095 002158 002064 002091 002120 002184 002164
002111 002122 002271 002165 002144 002201 002192 002160 002235 002249
002276 002248 002277 002205 002007 002002 001887 001933 002105 002287
002335 002608 002777 003040 003121 003036 003093 002903 002633 002498
0101
002219 002159 001930 001874 001816 001831 001702 001734 001703 001661
001567 001496 001468 001429 001519 001427 001472 001433 001407 001481
001490 001539 001445 001427 [illegible]413 001456 001375 001447 001427 001325
[illegible] 001321 [illegible] 001240 [illegible] 001224 [illegible]
[illegible] 000081 [illegible] 000074
[illegible]000075 000078 [illegible] 000063 000079 000073 000085 000061 000068 000062
000084 000053 000064 000055 000062 000062 000056 000052 000061 000078
0501
000064 000072 000051 000048 000064 000058 000066 000056 000068 000053
000047  LR-10     ZEP          413      9000.    600.0   1.75    02       09-18-81 3.0
```

Figure 10a. A sample sodium iodide spectrum as transmitted from the multichannel analyzer to the USGS computer. (The wavy line represents a break in the data.)

```
024413
009000
060000
175000
012817
0001
017924020686020447020398016922012428009019007326006493006303
006127006605006676006426006421006345006100005919005772005787
005735005904005821005920006234006361006522006837007505008690
010214011418011731010657008940007381005722004951004465004280
004205004165004231004082003838003879003821003925004276004583
004612004571004309003981003653003194002877002640002517002267
002266002232002226002095002158002064002091002120002184002164
002111002122002271002165002144002201002192002160002235002249
002276002248002277002205002007002002001887001933002105002287
002335002608002777003040003121003036003093002903002633002498
0101
002219002159001930001874001816001831001702001734001703001661
001567001496001468001429001519001427001472001433001407001481
001490001539001445001427[illegible]456001375001447[illegible]27001325
[illegible]
[illegible]
000075000078[illegible]000063000079000073000085000061000068000062
000084000053000064000055000062000062000056000052000061000078
0501
000064000072000051000048000064000058000066000056000068000053
LR-10     ZEP          413      9000.    600.0   1.75    02       09-18-81 3.0
```

Figure 10b. The same sample after editing to the proper format.

After the file has been edited, it can be accessed by the Alpha-M program only if the names file51 and file52 are added to the filename for detectors 1 through 8 and file53 and file54 are added to the filenames for detectors 9 through 16. The command to do this is "an filename file51 file52↓ "or "an filename file53 file54↓".

To interpret the spectral file using Alpha-M: "ear gam01↓" (enter absentee request) is typed to interpret detectors 1 through 4; "ear gam05↓" interprets detectors 5 through 8; "ear gam09↓" interprets detectors 9 through 12; and "ear gam13↓ " interprets detectors 13 through 16. These commands cause the programs to be run after normal working hours and the results may be obtained the following morning at the Computer Center in building 53. "dp filename" (dprint) causes a listing of the information on the cassette to be printed at the computer center and it may be picked up with the interpreted spectral data.

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