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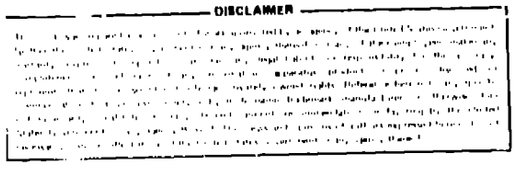

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A DUAL GERMANIUM DETECTOR SYSTEM FOR THE ROUTINE ASSAY OF LOW LEVEL TRANSURANICS IN SOIL

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ABSTRACT

As an outgrowth of previous work on soil radioassay, we have developed an automated assay system for determining the transuranic radionuclide content of soils, with particular interest in Pu. The system utilizes two commercial planar intrinsic germanium detectors in opposition. The large area of the detectors (2100 mm^2) and the thinness of the detector crystals (7 mm) permit sensitive analysis of the L x ray emission region of the transuranics (13-21 keV). With counting times of 5 hours, we obtain detection limits of <15 pCi/g for Pu and <0.1 pCi/g for ^{241}Am.

INTRODUCTION

Low level analyses of transuranic radionuclides are required in support of biological, ecological, and environmental research associated with nuclear facility operations. A photon spectroscopy system is described which is designed for the automated assay of transuranics in batch soil samples. The principal design criteria for this system were: a) minimum turnaround time for sample assay, b) little or no sample preparation, c) the ability to detect and quantify the concentrations of plutonium, americium, and uranium, and d) complete automation.

Analyses of low level transuranics in soil generally require costly, time-consuming chemical separation of radioactive material from the soil.[1] The separated fraction is then measured with high sensitivity alpha energy analysis or mass spectrographic methods. Direct photon spectroscopy of the soil sample offers a faster and less expensive alternative. Brauer et al[2] have measured ^{241}Am and Pu in soil by photon spectroscopy by using a Ge(Li) detector for measurement of the 60 keV γ ray from ^{241}Am and a Si(Li) detector for the measurement of the Pu/^{241}Am L_β x-ray ratio. Sherman et al[3] have described a Si(Li)-NaI(Tl) x-ray spectrometer for plutonium analysis in which

NaI(Tℓ) scintillators are operated in anticoincidence with a Si(Li) detector to reduce continuum in the L x-ray spectrum resulting from γ activity in the soil.

The system presented here is an outgrowth of the studies by West et al[4] in which a single crystal intrinsic germanium detector was used to detect Pu and ^{241}Am. The system, shown in Figure 1, utilizes two planar detectors of intrinsic germanium in opposition. The sample of soil, in a covered plastic dish, is positioned between the detectors for counting. Spectra in the energy range of 0-200 keV are accumulated from each detector by a microcomputer-based multichannel analyzer. The same microcomputer controls the automatic sample changer, analyzes the spectra, and prints the results on the computer terminal.

The sample preparation for this system is held to a minimum. After removal of sticks, stones, organic debris, etc., the soil is packed into plastic dishes 6.5 cm diameter x 0.5 cm deep. The dishes are covered and sealed. Packing density and drying of the samples are not critical so long as calibration standards of the same soil type are identically prepared.

DETECTORS

The detectors, manufactured by Princeton Gamma-Tech,[5] are single crystal planar detectors of intrinsic germanium, each mounted on a 30 l capacity liquid nitrogen dewar. Apart from the mounting geometry, the two detectors are essentially identical. The active area of each detector is 2100 mm^2 and the depletion depth is 7 mm. The windows on the detectors are 0.5 mm thick beryllium, 58 mm in diameter (≈2600 mm^2). This optimum window size was established by Monte Carlo calculations.[4]

The thin depletion depth of 7 mm ensures that the detectors are relatively insensitive to photons having an energy greater than about 200 keV. Figure 2 compares the detection efficiency of one of these detectors with that of an intrinsic germanium coaxial detector. This insensitivity to high energy photons is accompanied by reduced Compton scattering, thus reducing the continuum background in the 12-25 keV energy region (the region of the L x rays associated with the decay of transuranics). With depletion depths of less than 7 mm, energy resolution degrades rapidly due to capacitive effects. The energy resolution specifications of these planar detectors are 693 eV full-width at half-maximum (FWHM) at 6.4 keV and 787 eV FWHM at 122 keV. Although the energy resolution of so thin a detector is not as good as that of a smaller, thicker detector, the increased sensitivity for low energy

photons for these large area detectors was an overriding consideration. The resolution is still sufficient to resolve the L_α, L_β, and L_γ x rays of the transuranics, and the sensitivity at 186 keV is still sufficient to allow detection of uranium, as shown in the spectrum of uranium ore in Figure 3.

SYSTEM HARDWARE

The proposed use of this system is the unattended, automatic analysis of soil samples. For this purpose, a reliable sample changer which could be controlled by a computer was designed and constructed at this laboratory.[6] A stack of up to 23 plastic sample dishes can be analyzed in each run of the computer program. The stack loading sample changer, upon command from the computer, positions one sample at a time between the detectors, dropping the previous sample into a collection bucket. (See Figure 4.)

Because analysis of the spectra from the detectors was to be done by computer, and likewise control of the sample changer, a microcomputer-based multichannel analyzer (MCA) was selected as the heart of the system. The Model ND660 from Nuclear Data[7] utilizes a Digital Equipment Corporation[8] LSI-11 microcomputer. The same microcomputer which controls all of the MCA functions can, with minor modifications to Nuclear Data's hardware and software, also control the sample changer and allow a user-written analysis program to run concurrently with the MCA program.

SYSTEM OPERATION

In the normal operating situation, there are two programs running concurrently in the microcomputer. The "foreground," or high priority, program controls the MCA for spectrum acquisition and display. It, in turn, is controlled by the "background" program which performs the assay calculations and controls the sample changer. The manual controls of the MCA are disabled. The signals from the two detectors are directed, by an analog router, through a single analog-to-digital converter. The spectra from the two detectors are stored in two halves of a 4096 channel MCA memory (which is, in fact, random access memory of the microcomputer).

After the operator has entered the pertinent identification data for the samples to be analyzed, the background program causes the first sample to be placed between the detectors and spectra are accumulated for 1000 seconds. From the counting rate of the L_β x ray, the time necessary for sufficient statistics is determined (up to a maximum of 20,000 seconds) and,

if necessary, data acquisition is resumed. After the spectra are accumulated, they are summed (allowing for different energy calibration of the two detectors) into the first half of the MCA memory. A room background spectrum, stored on the computer's floppy disk, is loaded into the second half for subtraction.

The program scans a file on the floppy disk for a list of nuclides to be examined. For each γ ray or x ray of interest, the disk file contains the identity of the nuclide, the energy span of the peak, and the system sensitivity in (pCi/g)/(counts/sec). After subtracting sample and room background, the net counting rate in each energy region of interest and the concentration of the associated nuclide are determined. The results are printed in a table on the computer terminal, the next sample is moved into position, and the process is repeated.

ASSAY

With the exception of plutonium, each radionuclide is quantified on the basis of γ rays (e.g., 60 keV for ^{241}Am and 186 keV for ^{235}U/^{238}U mixtures). For low energy photons, like the L x rays, the mean free path in soil is about 1 mm, and only the surface layers of the sample are seen by the detectors. The counting rate is assumed to be proportional to the concentration of the nuclide in the soil. For higher energy photons, the mean free path is of the same order of, or larger than, sample thickness, and the counting rate is assumed to be proportional to the total amount of nuclide in the sample. For the sake of simplicity, a threshold of 65 keV was chosen to delineate the two approximations. (The errors introduced in the middle energy region by this simplification seem to be considerably less than those inherent in the counting statistics.)

The plutonium concentration is determined from the counting rate of the L_α, L_β, and L_γ x rays after a correction is made for the ^{241}Am and U contributions, using predetermined fractions (obtained from ^{241}Am and U soil standards) of the 60 keV and 186 keV peaks. A summary of the assay for Pu, ^{241}Am, and U for each sample is printed below the table of nuclides described above.

CALIBRATION

The accuracy of this soil analysis system is critically dependent upon the use of well-prepared calibration standards. The absorption of the L x rays by the sample itself (and thus the ratio of L x-ray counting rate to γ-ray

rate for ^{241}Am and U) is highly matrix dependent. Well characterized, preferably spiked, standard of the same type of soil to be analyzed must be available to determine the sensitivity of the system for each radionuclide and each soil type.

SUMMARY

A photon spectroscopy system for the automated analysis of transuranics in soils has been described. The system is now in routine use. Detection limits (based upon background statistics for individual samples) are less than 15 pCi/g for Pu and less than 0.1 pCi/g for ^{241}Am. A reportable detection limit for U awaits measurements of actual samples with detectable amounts of that element.

ACKNOWLEDGEMENTS

I would like to thank Donald A. Close for performing detector efficiency measurements. I would also like to thank George Trujillo of the LASL Environmental Studies Group for preparation of soil standards and samples. I would like to express particular appreciation to Faustin Trujillo for mechanical design and fabrication of the automatic sample changer.

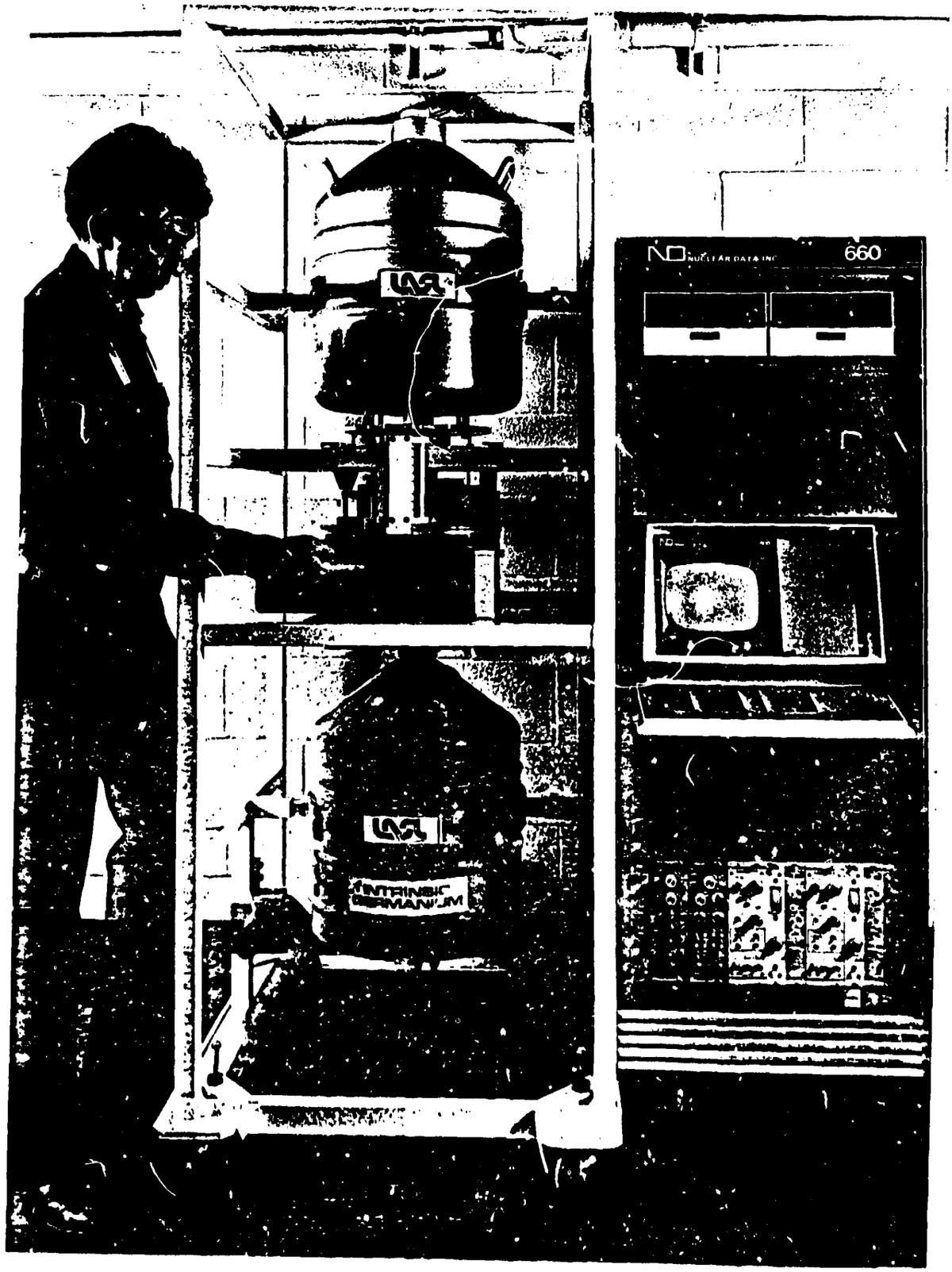

Figure 1.

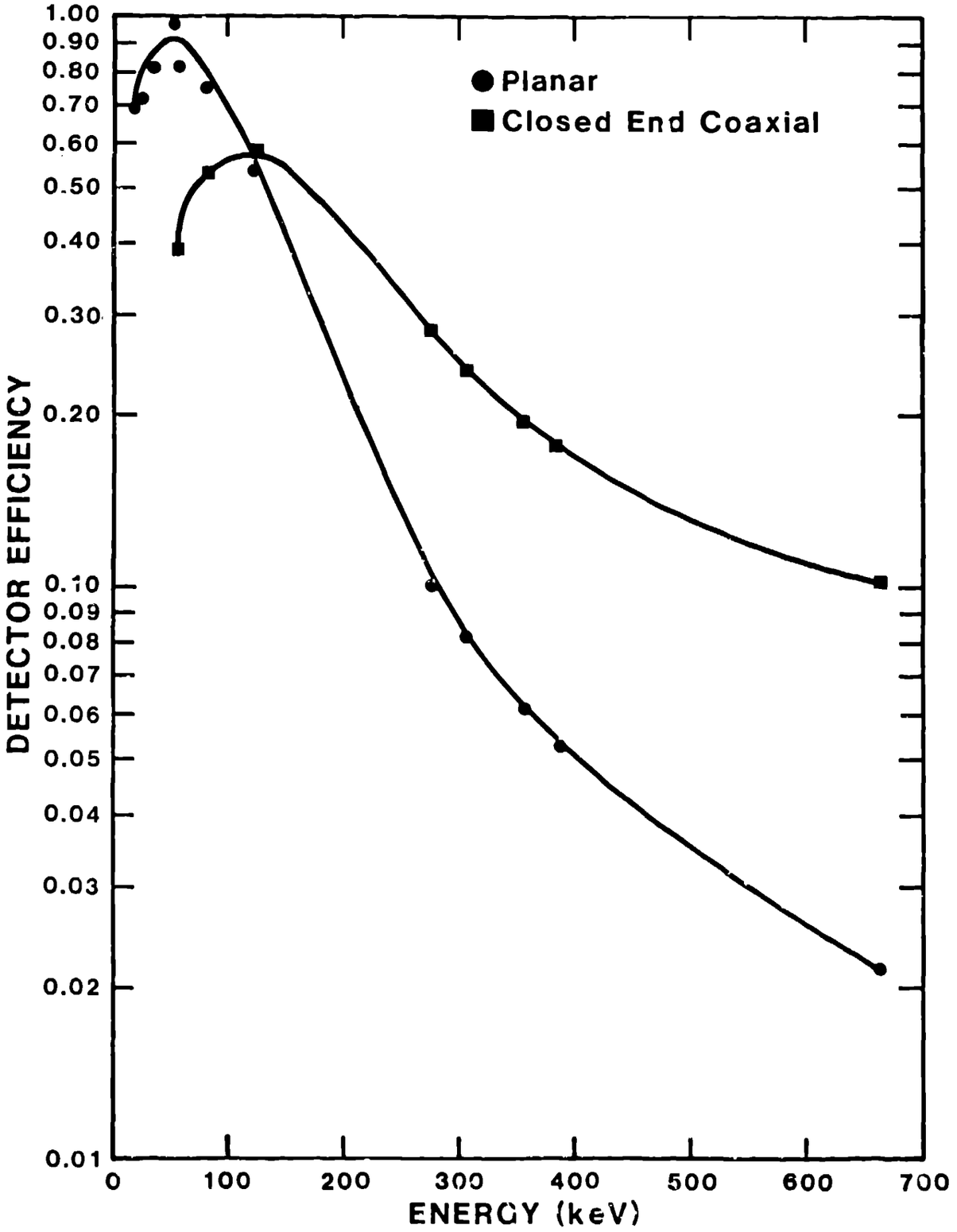
1.00
0.90
0.80
0.70
0.60
0.50
0.40
0.30
0.20
0.10
0.09
0.08
0.07
0.06
0.05
0.04
0.03
0.02
0.01
DETECTOR EFFICIENCY
Planar
Closed End Coaxial
0
100
200
300
400
500
600
700
ENERGY (keV)

Figure 2.

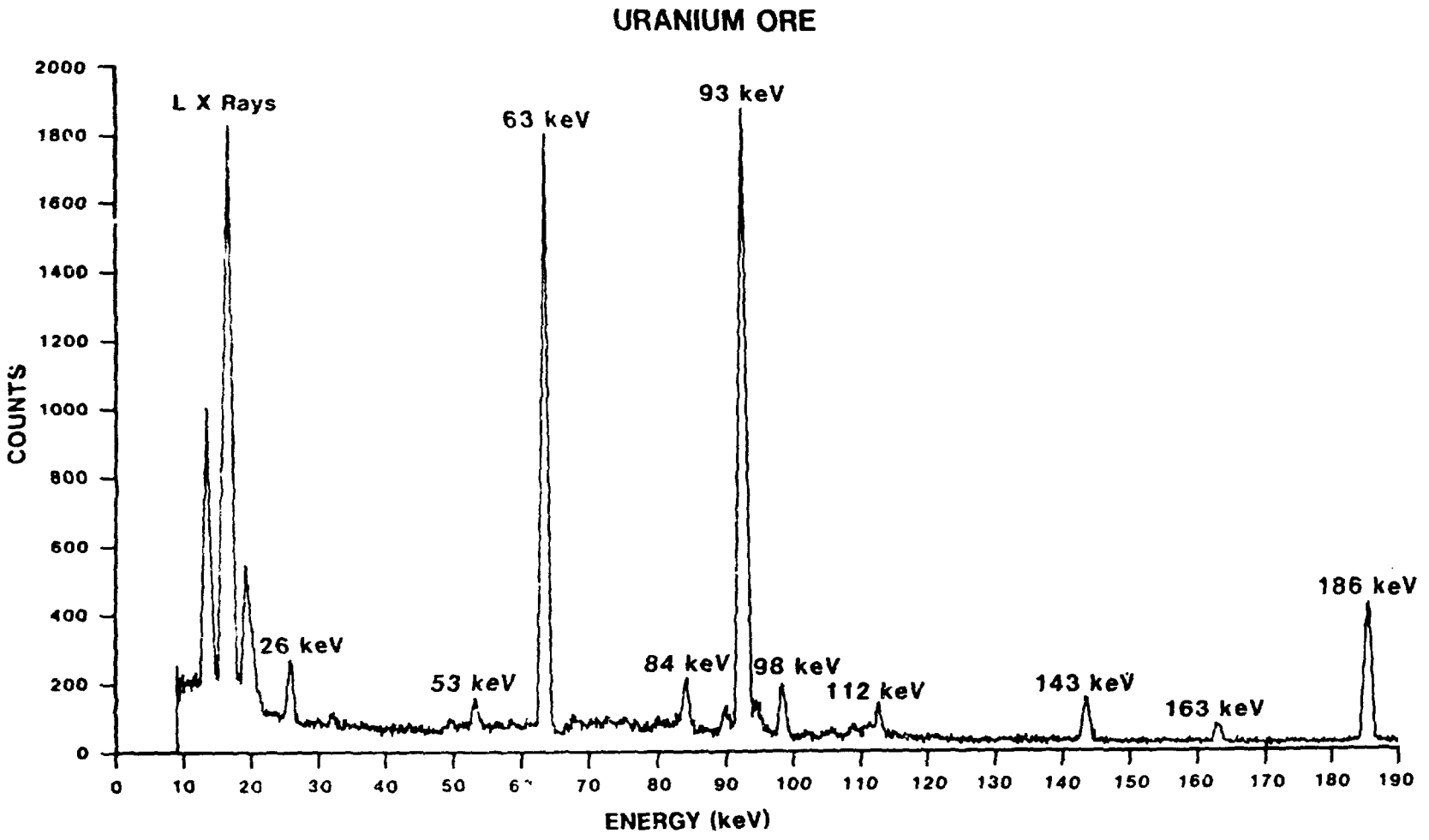
URANIUM ORE
L X Rays
63 keV
93 keV
186 keV
26 keV
53 keV
84 keV
98 keV
112 keV
143 keV
163 keV
COUNTS
ENERGY (keV)
2000
1800
1600
1400
1200
1000
800
600
400
200
0
0
10
20
30
40
50
70
80
90
100
110
120
130
140
150
160
170
180
190

Figure 3.

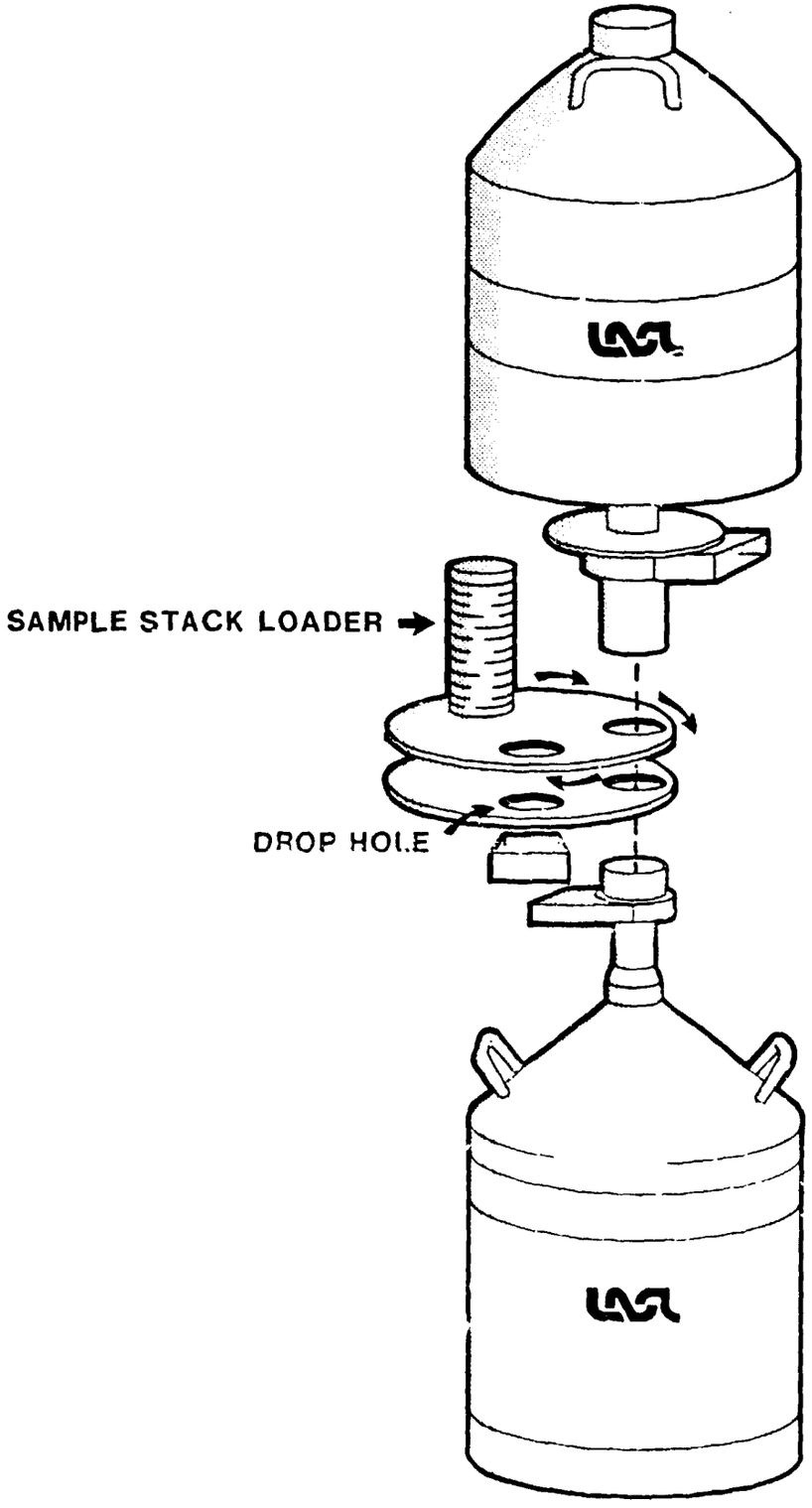

Figure 4.

REFERENCES

1. R. J. Rudr "Plutonium: A Review of Measurement Techniques for Environmental Monitoring," IEEE Trans. on Nucl. Sci, NS-21, 430 (1974).

2. F. P. Brauer, J. M. Kelley, R. W. Goles, and J. E. Fager, "Measurement of Environmental ^{241}Am and the Pu/^{241}Am Ratio by Photon Spectroscopy," IEEE Trans. on Nucl. Sci., NS-24, 591 (1977).

3. I. S. Sherman, M. G. Strauss, and R. H. Pehl, "Measurement of Trace Radionuclides in Soil by L X-Ray Spectrometry," IEEE Trans. on Nucl. Sci., NS-27, 695 (1980).

4. L. West, C. J. Umbarger, and T. Dempsey, "A Germanium Detector System for the Detection of Transuranics at Low Activity Concentration in Soil," Proceedings of the Eleventh Midyear Topical Symposium of the Health Physics Society on Radiation Instrumentation, San Diego, CA, January 16-19, 1978, page 564.

5. Model 2107, Princeton Gamma-Tech, P.O. Box 641, Princeton, NJ 08540.

6. F. Trujillo and J. M. Crowell, to be published.

7. Model ND660, Nuclear Data, Inc., Golf & Meacham Rds., Schaumburg, IL 60196.

8. Digital Equipment Corporation, Maynard, MA 01754.

FIGURE CAPTIONS

Figure 1. Automated Soil Analysis System.

Figure 2. Efficiency of 2100 mm^2 x 7 mm planar and 1400 mm^2 x 34 mm coaxial intrinsic germanium detectors.

Figure 3. Spectrum of uranium ore taken with 2100 mm^2 planar intrinsic germanium detector.

Figure 4. Detail of sample changer mechanism.