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NONDESTRUCTIVE DETERMINATION OF RADIONUCLIDES IN LUNAR SAMPLES USING A LARGE LOW-BACKGROUND GAMMA-RAY SPECTROMETER AND A NOVEL APPLICATION OF LEAST-SQUARES FITTING*

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Abstract

Dual-parameter gamma-ray spectrometer systems with large-volume NaI(Tl) crystals and ultra-low backgrounds have been used for the non-destructive determination of K, Th, U, and cosmic-ray produced radionuclides in 60 lunar samples. The total weight of samples measured with these systems is ∿28 kg, and the individual sample weights varied from 2 to 2300 g. Samples from Apollo 11, 12, 14, 15, and 16 missions have been measured. Operation of the spectrometers in a coincidence mode and analyzing singles- and coincidence-spectra permits the simultaneous determination of 8-10 radionuclides in each lunar sample.

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1. Spectrometer Design and Operation

The spectrometers contain two NaI(Tl) detectors, each 23 cm in diameter and 13 cm long, with 10 cm pure NaI light guides. The detectors are operated in a coincidence mode along with a 30 cm thick plastic scintillator mantle in anticoincidence with the NaI(Tl) detectors for background reduction and improvement in peak-to-total response. Further background reduction is achieved by surrounding the entire assembly with a lead shield 20 cm thick. Figure 1 shows a cross-section of the final detector assembly used in the lunar sample analysis program. Considerable development effort preceded the final design. For example, identical detectors (23-dia x 13-cm length) were constructed with and without the NaI light guides. It was found that the detectors with light guides had a background counting rate that was 2.2 times lower than the detector without the light guide. This reduction is achieved because of the attenuation of ^{40}K radiations from the phototubes by the light guide (even the metal jacketed RCA 4521 phototubes contain some glass). Low-background lead was carefully selected for the shield construction (24 metric tons) by surveying samples of lead bricks prepared from virgin ore deposits and comparing background levels with old lead that had been purified by melting. Lead for the shield construction was selected on the basis of its gamma-ray emission rate in the energy range 80-1400 keV. As a practical approach, it was decided to compare the gamma-ray emission of the test sample (19 lead bricks 5 x 10 x 20 cm surrounding a 13 x 10-cm NaI(Tl) detector in close proximity) with that of the old lead as a reference material. It should be noted that the old lead was available only in a quantity large enough for the test. The lead used in the final construction had a gamma-ray emission rate 1.02 times the standard, while two other contemporary samples had 1.43 and 3.51 times as great a rate as the standard.

Data acquisition in these systems is accomplished with dual 12-bit analog-to-digital converters and a coincidence-anticoincidence logic circuit interfaced to a 4096-channel analyzer or an 18-bit digital computer having 16,384 words of core memory. Coincident gamma-ray interactions are recorded in a 64 x 64 channel matrix in the case of the wired analyzer or in a 128 x 128 channel folded matrix for the computer based system.

Quantitative radionuclide determination from the various collected spectra is accomplished with ALPHA-M[1]), a program for least-squares resolution of gamma-ray spectra. The accuracy of quantitative determinations by the least-squares technique is strongly dependent on the similarity of the standard response function of each radionuclide to the corresponding response in the unknown sample. Accordingly, it is necessary to collect responses from calibration standards in the exact geometry and electronic density as that of the lunar samples. Electrolytically reduced iron powder was found to be a suitable dispersing medium for the 8-10 standards needed for each analysis. Replicas of rocks are constructed of 0.08 mm aluminum foil shells hand-formed from rock models, reinforced with epoxy cement and filled with a standard radionuclide-iron powder mixture. A collection of responses for each set of replicas is made into a library for final analysis of the lunar samples in the least-squares program.

The 4096- or 8192-channel gamma-gamma coincidence matrices contain many zero-value channels and are too large to analyze conveniently by least-squares techniques. Statistical considerations concerning the large number of zero-value channels and the desire to condense the matrix so that gain-shift options could be employed in the least-squares program resulted in a novel scheme of dividing the coincidence matrix into several zones and summing planes prior to application of the least-squares program. The folding and summation scheme results in the condensation of 8192-channel spectra to 256 channels and that of the 4096-channel spectra to 168 channels.

2. Results

Efficiency factors (cpm/dpm for cosmogenic radionuclides and cpm/mg for the primeval radioelements) were calculated for one of the standard geometry conditions used in the analysis of 100-g samples of lunar soil. These factors are shown in Table 1 for a 5 cm detector separation with the sample covering 46 cm^2 midway between the detectors. The high coincidence counting efficiency achieved in this system is due to the use of a large fraction of the entire coincidence matrix from the folding and plane summation technique by the least-squares program. The usual fit region for the analysis of the singles spectra encompasses the energy range 0.4-3.6 MeV. The background counting rate for the summed singles output from both detectors (10,400 cm^3 active volume NaI) is 80 cpm. The usual fit region for the least-squares analysis of the folded and summed coincidence spectra is made up of four segments: 1) those events in the energy region 0.36-2.94 MeV from one detector that are in coincidence with events in the range 0.12-0.30 MeV in the other, 2) those in the energy region 0.36-3.18 MeV in coincidence with 0.36-0.72 MeV in the other detector, 3) those in the energy region 0.96-3.30 MeV in coincidence with 0.78-0.96 MeV in the other detector, and 4) those in the energy region 1.02-2.70 MeV in coincidence with 1.02-1.62 MeV in the other detector. The background counting rate for this entire coincidence region is 3 counts per minute.

3. Conclusions

The internal consistency of the experimental procedures were checked with the help of standard radionuclide test mixtures incorporated in lunar sample replicas. Amounts taken and found agreed in each case within the limit of

Table 1

Efficiency values (cpm/dpm) in the lunar sample spectrometer for the analysis of 100-g samples of soil with a detector separation of 5 cm.

Radionuclide	Singles efficiency	Coincidence efficiency
^{22}Na	7.5%	21.5%
^{26}Al	9.8%	20.3%
^{46}Sc	18.2%	13.2%
^{48}V	70.9%	16.9%
^{54}Mn	36.9%	1.4%
^{56}Co	7.2%	11.0%

Primeval Radioelement*	Singles sensitivity	Coincidence sensitivity
K	0.63 cpm/mg	0.003 cpm/mg
Th	78.9 cpm/mg	24.4 cpm/mg
U	154 cpm/mg	59.9 cpm/mg
Background	80 cpm	3.0 cpm

*Terrestrial isotopic composition.

counting statistics, usually 2 to 5%. A statistical analysis of 28 determinations of an Apollo 12 soil sample from various laboratories[2]) showed that our analyses of that sample for K, Th, and U fall within an average deviation from the mean of ± 2.6%, which is smaller than our reported errors[3]).

The combination of two computer techniques, namely, that of plane summation and condensation of the matrix coupled with the least-squares analysis, has provided a powerful technique for resolving a large matrix in the multidimensional gamma-ray spectrometer. With the ultra-low-background system and computer techniques, we have demonstrated the ability to determine quantitatively mixtures containing 8-10 gamma-emitting radionuclides at the 10-100 dpm/kg level, even in the presence of thorium and uranium at the 3-10 ppm level.

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Fig. 1. Design of the low-background gamma-ray spectrometers for the analysis of lunar samples. The symbol NE-102 refers to a plastic scintillator material.

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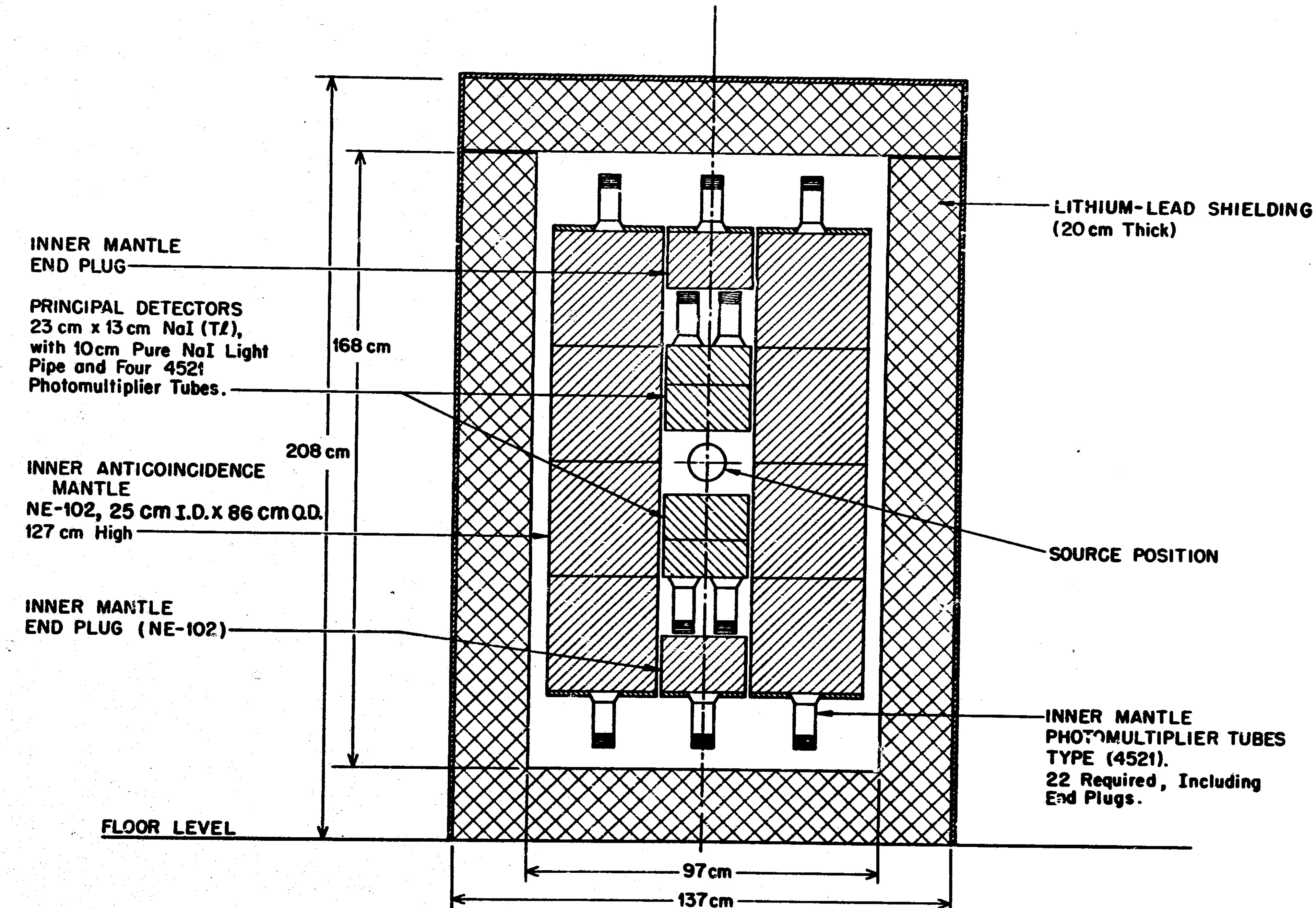

Design of the Anticoincidence Gamma-Ray Spectrometer