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Use of NaI (Tℓ) and germanium detectors for in situ γ-ray spectral monitoring of boreholes at nuclear waste-disposal sites

by

W. S. Keys, F. E. Senftle, and A. B. Tanner

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ILLUSTRATIONS

USE OF NaI(Tl) AND GERMANIUM DETECTORS FOR IN SITU γ-RAY SPECTRAL MONITORING OF BOREHOLES AT NUCLEAR WASTE-DISPOSAL SITES

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Abstract.--Gamma spectrometry in boreholes has been used experimentally for the in situ identification of radioisotopes migrating from several radioactive waste-disposal sites. Although the technique has been successfully demonstrated using sodium iodide (NaI) detectors, both the instrumentation and techniques of data analysis need improvement. Sodium iodide detectors have high counting efficiency but poor energy resolution. In contrast, semiconductor detectors have excellent resolution but low efficiency. This report describes a field study comparing results with the two types of detectors at Oak Ridge, Tennessee. A high-purity planar germanium detector and a 3.2 x 10.2 cm NaI crystal were used for the comparison study. Both enabled the unambiguous identification of the naturally occurring radioisotopes and cobalt-60 and cesium-137. The germanium detector, however, permitted the identification of a number of additional radioisotopes that emit gamma radiation of lower energy. Neither of the systems is yet calibrated to permit quantitative radioisotope analysis in boreholes. The germanium detectors show promise for borehole monitoring but several factors will inhibit routine field use in the immediate future. These include high cost, operation at cryogenic temperatures, standard logging cable, and long counting times. Additional studies are underway to improve the utility of both types of systems.

INTRODUCTION

Over the past thirty years, it has been common practice to dispose of low-level radioactive waste by burial in shallow trenches. At such sites, radioactive contaminants can be leached or removed from the waste materials by water that is initially present in the trenches, or from rainfall or surface runoff that later infiltrates the trenches. Unless this water is pumped from the trenches, the radioactive material can be carried away from the trenches in ground water, and later discharged into river systems. It is important, therefore, to monitor the ground water in and around these waste burial sites so that remedial measures may be initiated if unacceptably high concentrations of radionuclides are found. Although water samples taken from streams, wells, and trenches ordinarily are analyzed in the laboratory, it is equally or more important to measure in situ the radioactivity of subsurface water intersected by boreholes. Spectral rather than total radioactivity measurements made in boreholes may provide immediate identification of artificial radioisotopes as well as information on their vertical distribution. Identification can also be made of contaminants outside the casing where they cannot be sampled. Spectral borehole measurements also provide radionuclide identification in those burial sites where there are no records of the type of waste that has been interred.

DESCRIPTION OF DETECTORS

The identification of radioactive isotopes in a borehole has been done by Keys, Eggers, and Taylor (1977) with an NaI(Tℓ) scintillation detector mounted in a sonde. ^{60}Co, ^{134}Cs, and ^{137}Cs have been identified to date. Although this type of detector is relatively efficient and adequately sensitive to detect the total gamma activity, it does suffer from poor energy resolution.

The use of a semiconductor type detector is an alternative method which can be used to obtain good energy resolution. Voropaev, Martynov, and Sulin (1974) have compared the energy resolution, efficiency, and peak-to-Compton amplitude ratios of NaI(Tℓ) and Ge(Li) semiconductor detectors, and found both to have comparable sensitivities for monochromatic gamma radiation containing only one line; however, for congested spectra containing many close-spaced lines, the Ge(Li) or high purity germanium semiconductor is the only detector which has adequate resolution.

Nielson, Thomas, Wogman and Brodzinski (1976) have compared several methods of measuring spectra of low energy γ-rays in the laboratory. Their results also indicate that a high-purity germanium detector has resolution far superior to that of NaI(Tℓ) detectors for the identification of plutonium and americium on soil surfaces and pond bottoms.

Subsequently Nielson, Wogman, and Brodzinski (1977) described the use and adaptation of Ge(Li), high purity germanium, NaI(Tℓ) and plastic phosphor detectors to the in situ measurement of subterranean gamma-ray emitting radionuclides. Although they did not present any field spectra, they discussed the application of the technology to the in situ measurement of radionuclides leaked from underground fuel waste storage tanks.

Use of a high-purity germanium detector in a borehole at a depth of a hundred meters or more, however, presents the following problems:

1. The device must be operated near liquid nitrogen temperatures (-196°C)
2. Low-energy X-ray and γ-rays are more strongly absorbed by the borehole wall, ground water, and casing than are the γ-rays of higher energy, resulting in a lower counting rate and smaller effective sample size.
3. Because the detector must be mounted in a watertight sonde, the walls of the sonde must be thin enough to minimize absorption but thick enough to maintain the mechanical integrity of the sonde.
4. The counting efficiency of a high-purity germanium detector is much less than that of a NaI (Tℓ) detector; hence longer counting times must be used to obtain comparable statistics.
5. A germanium detector must be operated with a cable having a coaxial member instead of a standard armored cable.

A comparison of some salient features of the two detectors is given in table 1.

Table 1 ------- Near Here

Table 1.-Advantages and disadvantages of low energy Na(Tℓ) and germanium detectors in borehole sondes

	NaI(Tℓ)	Thin planar germanium
Maximum operating temperature*	<373 K	<100 K
Environmental thermal shock	Poor	Good
Resolution at 122 keV	15 keV	0.8 keV
Efficiency	5-20 percent	~5 percent of NaI(Tℓ)
Cable	Any standard logging	Coaxial member
Cost	Less than $1000	From $5,000 to $10,000

*Operating temperature of detector proper. In an insulated sonde, a NaI(Tℓ) detector may be used in ambient temperatures of 450 K. The housing of the Ge sonde should permit operation to about 325 K without additional insulation, which could extend the range upward substantially.

Senftle, Moxham, Tanner, Boynton, and Philbin (1976) described a high-purity planar germanium detector mounted in a borehole sonde that was developed for uranium exploration. A cryogenic system described by Boynton (1975) was incorporated into the sonde to keep the germanium cold. The detector was surrounded by a 0.76 mm aluminum window to reduce low-energy γ-ray absorption. Although this particular sonde is somewhat lacking in sensitivity, it was felt that it could be used to compare results with a conventional NaI(Tℓ) type sonde in a nuclear-waste burial site.

THE TEST SITE

Burial ground 5 at Oak Ridge National Laboratory, Tennessee, was chosen to conduct the comparison between the two detection systems. This site, which was operational between 1958 and 1973, is described by Duguid (1975) as being similar to a sanitary landfill. The waste was simply placed in parallel, unlined trenches and covered with approximately 0.6 m of soil (fig. 1).

Figure 1.--NEAR HERE

The site is bordered on the west by White Oak Creek and on the south and east by the Melton Branch, a tributary to White Oak Creek. Rainfall and runoff infiltrate the trenches and mingle with the contaminated waste. Ground water moving below the site is thought to discharge to both streams. Shallow observation wells utilizing PVC plastic casing were drilled at strategic locations in and around the trenches to obtain data on water levels.

THE EXPERIMENTAL PROCEDURE AND RESULTS

The scintillation sonde consisted of a 3.2 x 10.2-cm NaI(Tℓ) crystal mounted in a 5-cm diameter sonde. A conventional preamplifier, linear amplifier, and power supply were also mounted in the sonde. The sonde was connected by a standard 4-conductor cable to the associated electronics, readout, and recording equipment in a logging truck. The high-purity germanium detector and sonde have been described previously (Senftle and others, 1976).

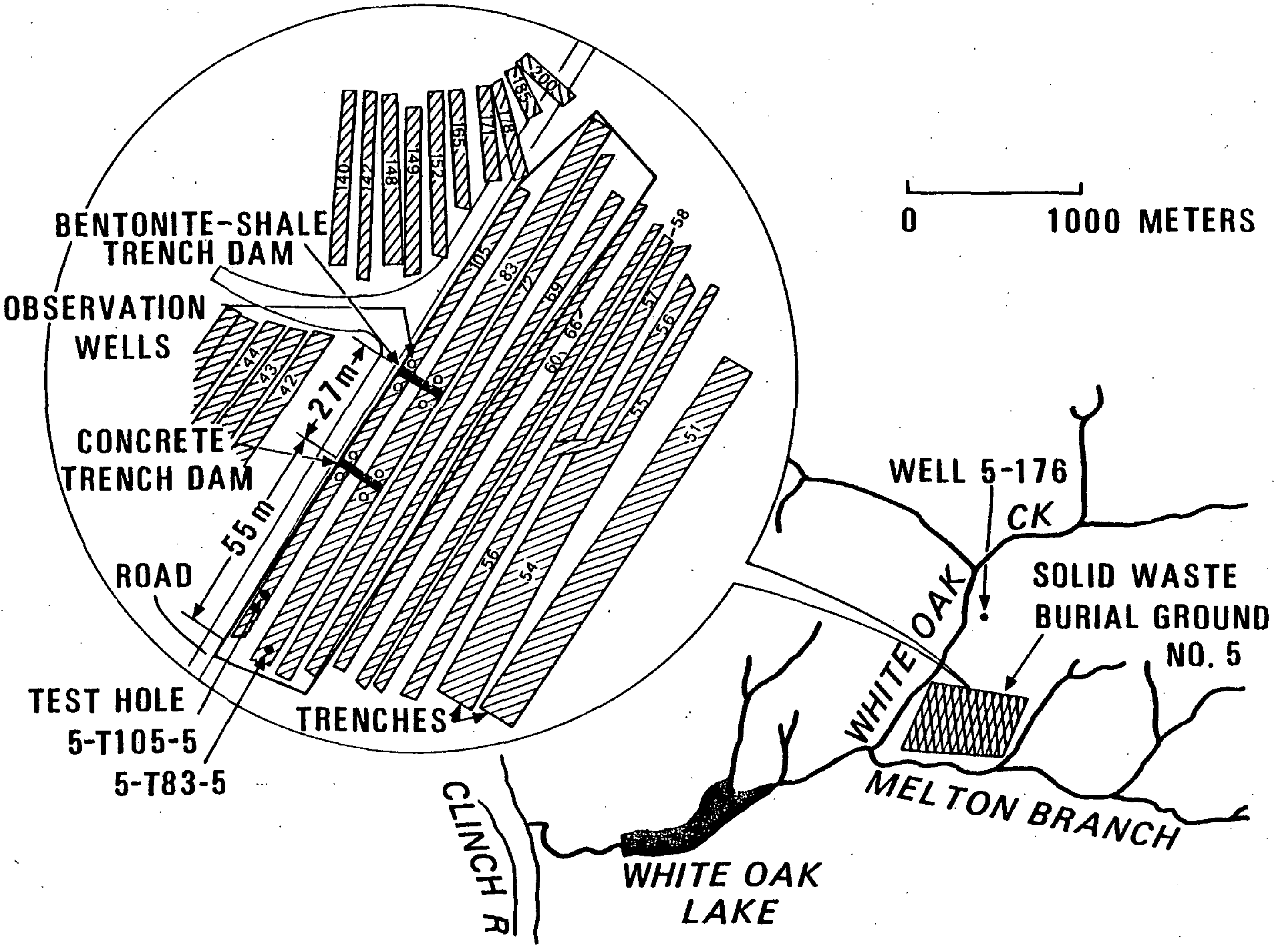

Figure 1.--Sketch showing location of nuclear waste-disposal site at Oak Ridge National Laboratory and an expanded view of the trench system.

In most of the boreholes examined, a complete log was first made with the scintillation sonde to determine the depth at which highest radioactivity occurs. Figure 2 shows typical spectrum with this instrument

Figure 2.—NEAR HERE

accumulated for 50 seconds at a depth of 1.74 m in well 5-T83-5, located in the end of a trench. The peaks at 661, 1,171, and 1,332 keV are ^{137}Cs and the two lines of ^{60}Co, respectively. The resolution of the detector was inadequate to permit identification of the many other artificial nuclides. The naturally occurring radionuclides are also presented in this spectrum, but are masked by the high activity and scattered radiation from the artificial radionuclides. The spectrum in figure 3 shows peaks

Figure 3.—NEAR HERE

from the natural radioisotopes and ^{137}Cs at a depth of 43.65 m in hole 5-176. The prominent peaks at 662 and 1,461 keV are ^{137}Cs and ^{40}K. The poorly developed peaks between 1,600 and 2,800 keV are from the natural uranium and thorium series. Note the poor statistics at these higher energies for a 1000-second counting period.

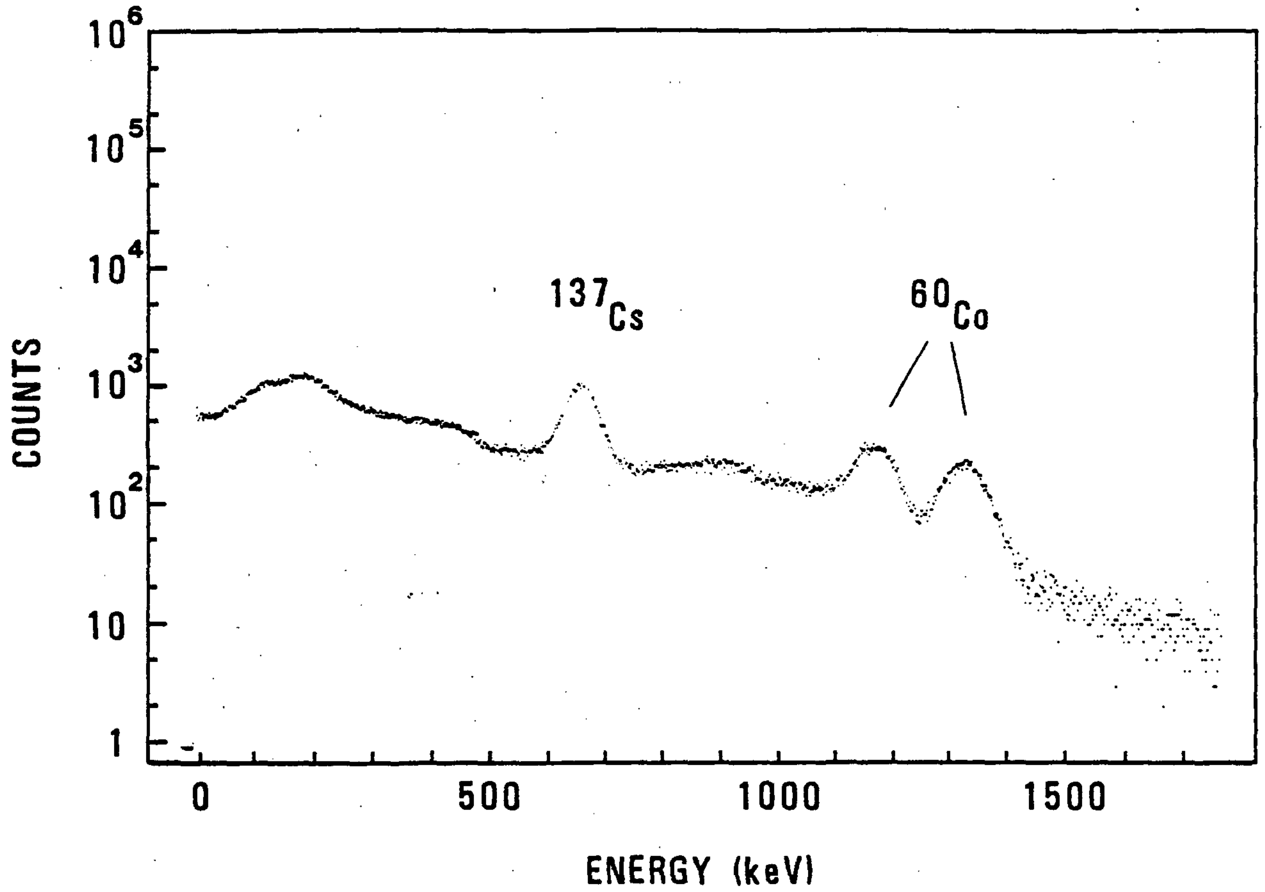

FIGURE 2.-- Spectrum taken 1.74 m below the surface in borehole 5-T83-5 using a NaI(Tℓ) detector.

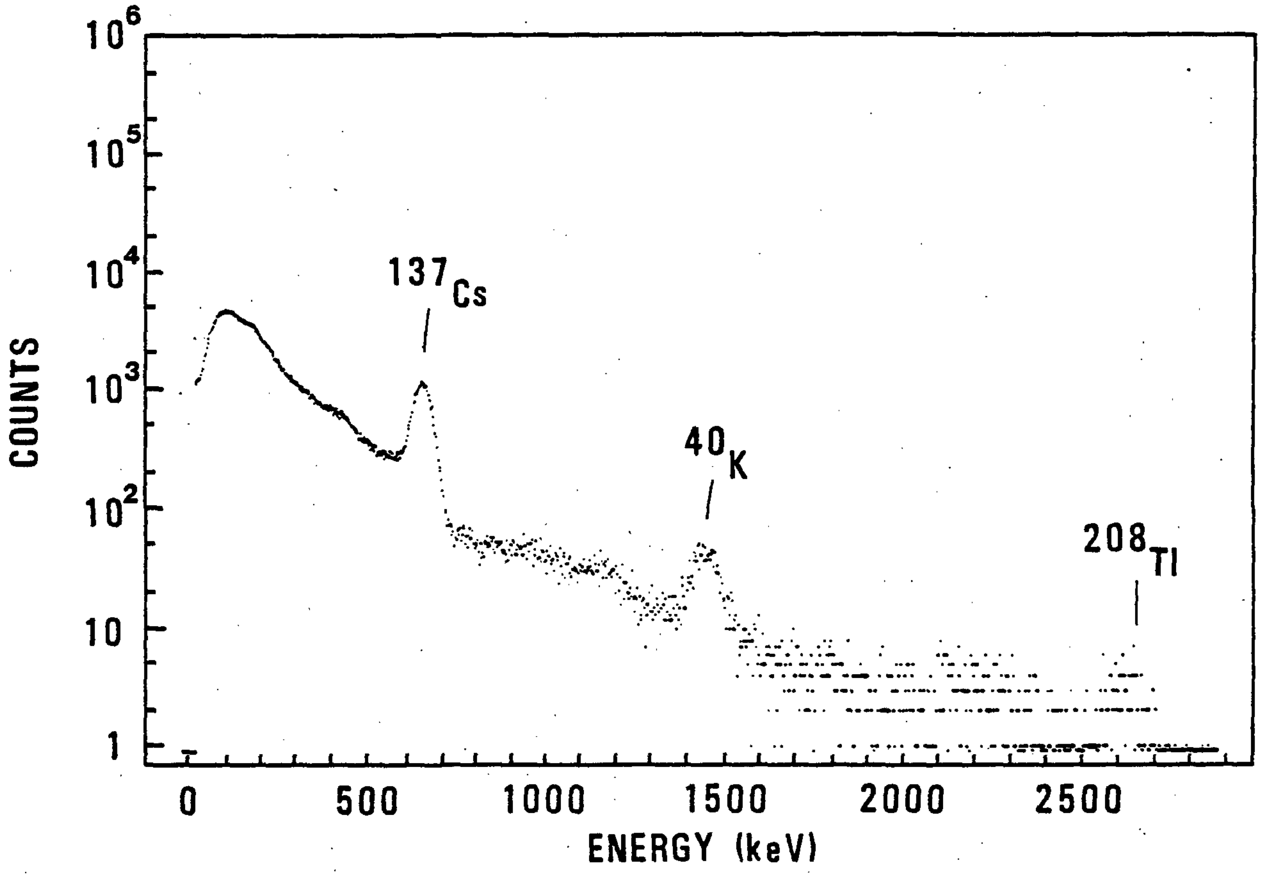

FIGURE 3.-- Spectrum taken at 43.65 m below the surface in borehole 5-176 using a NaI(Tℓ) detector.

Figure 4 shows a spectrum taken with the high-purity germanium

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Figure 4.--NEAR HERE

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detector in hole 5-T83-5 at a depth of 2.27 m. In addition to the ^{137}Cs and ^{60}Co lines, there are several smaller peaks about 300 keV which are slightly above 2.4 standard deviations. Many of these peaks are associated with members of the natural radioactive series. The small germanium detector used has a much lower efficiency at these higher energies and will therefore require a longer counting period than with a NaI(Tℓ) detector to obtain comparable statistics. Below 300 keV, six lines of ^{243}Cm and the 74.8-keV line of ^{243}Am can be seen. No attempt was made to make quantitative measurements in these initial tests. Calibration of the borehole sonde will have to be made in a special facility if further work of this nature is to be done in the future.

The radionuclide distribution varies sharply from depth to depth within the borehole. For example, the high-resolution spectrum taken at 3.61 m in hole 5-T105-5 and shown in figure 5 is quite different from

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Figure 5.--NEAR HERE

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that shown in figure 4 taken a few meters away. The distribution probably results from the heterogeneous nature of the waste placed in the trench, and to deposition of radionuclides leached from the waste at different water levels in the trenches. The relatively abrupt changes in radionuclide

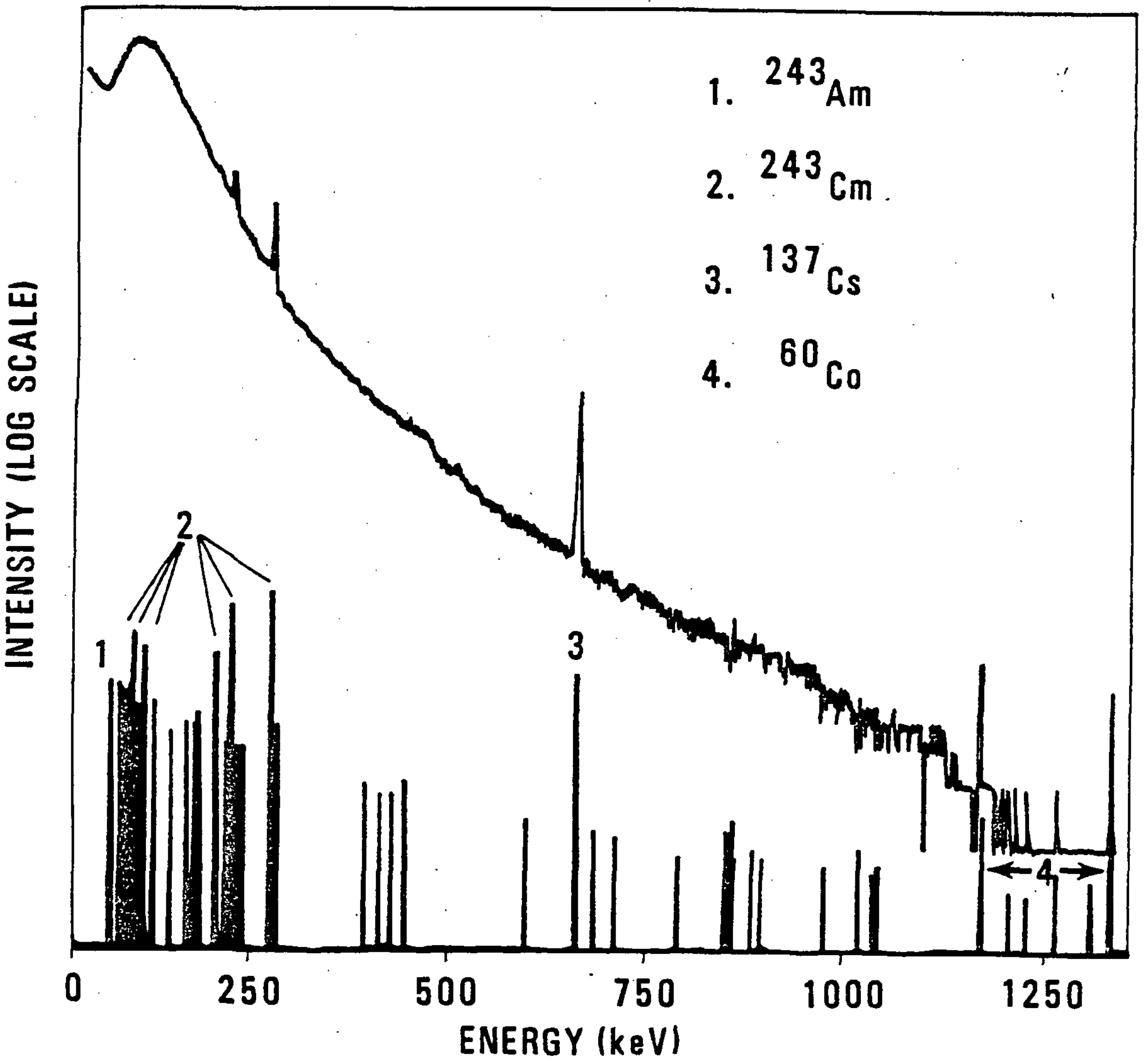

Figure 4.--Spectrum taken 2.27 m below the surface in borehole 5-T83-5 using a high-purity planar germanium detector. Reduced data are above a standard deviation of 2.45 and the peak identification numbers are: (1) ^{243}Am, (2) ^{243}Cm, (3) ^{137}Cs, and (4) ^{60}Co.

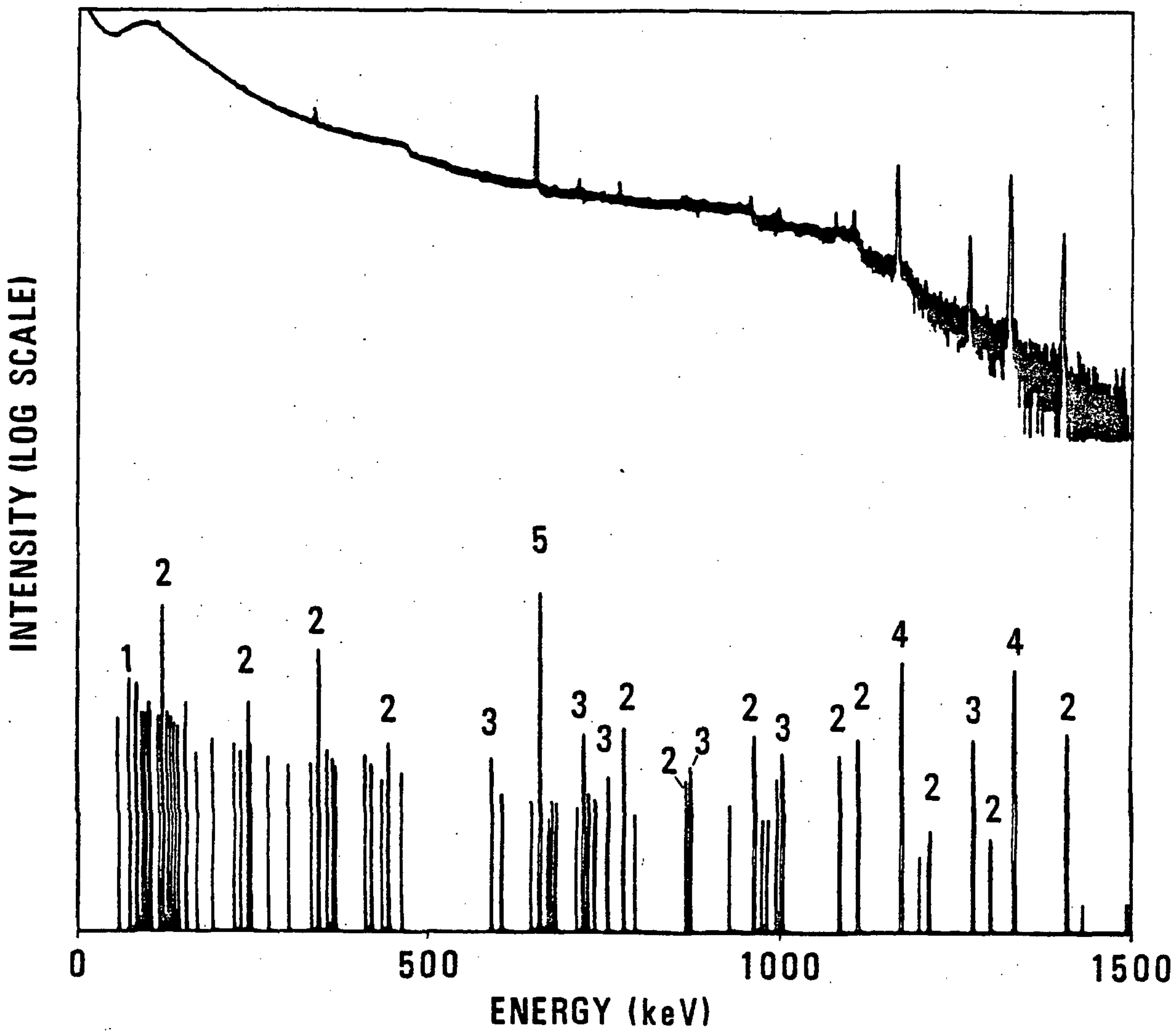

Figure 5.--Spectrum taken 3.61 m below the surface in borehole 5-T105-5 using a high-purity planar germanium detector. Reduced data are above a standard deviation of 2.45 and the peak identification numbers are (1) ^{243}Am, (2) ^{152}Eu, (3) ^{154}Eu, (4) ^{60}Co, and (5) ^{137}Cs.

assemblage are reflected in significant changes in spectral composition as some of the wells are logged. Spectra accumulated at six positions within hole 5-T105-5 provide an example. Figure 6 shows how the distribution of five of the nuclides changed with depth, and table 2 gives a list of the nuclides having the most intense lines in the spectra. Because of the unusual suite of nuclides present neutron logs were run in several of these holes. Although discrimination against the very high gamma background was a problem, it is not thought that neutrons are present.

Figure 6.--NEAR HERE.

Table 2.--NEAR HERE.

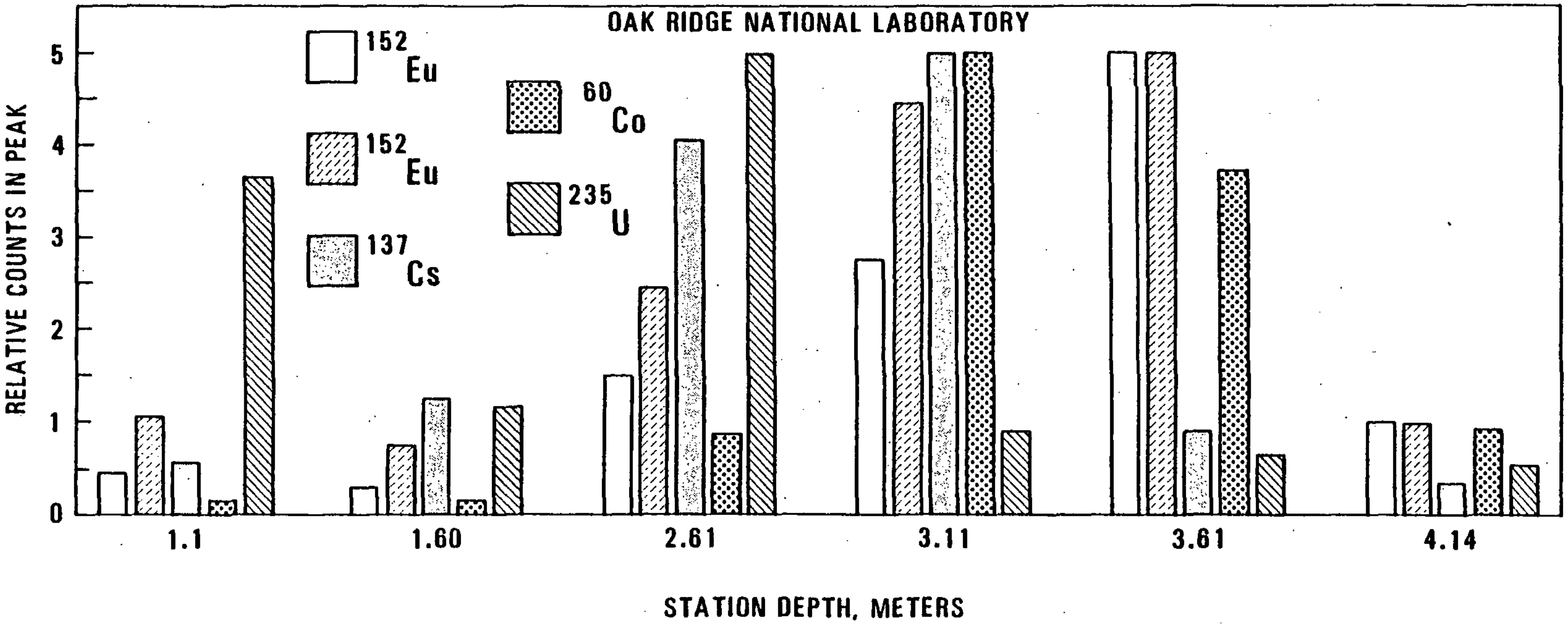

Figure 6.-- Bar graph showing the relative concentrations of five radionuclides with depth in borehole 5-T105-5 as determined with the germanium detector.

Table 2.- Counts normalized to 1200 s accumulation time in selected peaks of a given radionuclide at six depths in borehole 5-T105-5 with the germanium detector.

	Energy	T1/2	Depth (meters)					
Nuclide	(keV)	(yr)	1.11	1.6	2.61	3.11	3.61	4.14
^{243}Cm	278	30	108	-	716	740	67	*181
^{249}Cf	388	352	143	44	424	156	67	26
^{243}Am	75	7400	1606	534	370	1724	696	373
^{241}Am	60	458	1481	127	-	-	300	100
^{237}Np	86.5	2.14x10^{6}	789	439	1562	2136	588	*477
^{233}Pa	312	7.5x10^{-2}	152	131	322	568	86	28
^{235}U	163	7.1x10^{8}	763	242	1044	188	131	*106
^{234}U	53	2.48x10^{5}	207	352	682	-	239	100
^{152}Eu	344	12.7	125	81	400	724	1308	255
^{154}Eu	123	16	833	556	1798	3268	3649	1058
^{60}Co	1173	5.263	42	45	234	1312	978	235
^{137}Cs	661	30	3096	6018	22,882	26,836	4850	1645
^{178N}Hf	426	10	90	47	318	228	-	-
^{101}Rh	326	3	-	29	286	-	-	-

*Peak energy was slightly high or low (<^{2}eV) compared to listed value.

CONCLUSIONS AND RECOMMENDATIONS

These results show that a germanium detector permits identification of a greater number of low-energy radionuclides than does a NaI(Tℓ) detector. Development is required for both types of detectors to enable them to be used for quantitative analysis. A large tank containing known concentrations of radionuclides could be used to calibrate the sondes. Concentrations of ^{60}Co, ^{134}Cs, and ^{137}Cs have been calculated from spectra recorded with the NaI(Tℓ) detector using concentrations of naturally occurring radioisotopes for calibration (Eggers, 1976). The germanium detector used in the present experiments was small (200 mm^2). The sensitivity of the germanium sonde could be enhanced by increasing the surface area and perhaps by using tandem detectors in the same sonde. As the sonde would be used only at shallow depths, the sensitivity at the low energies could also be enhanced by using a thinner window fabricated from beryllium instead of aluminum. The sonde could then be used to detect energies as low as 30 keV, which would allow one to measure the ^{129}I at 40 keV and other long-lived, low-energy gamma emitters.

In addition to monitoring nuclear waste-disposal sites, the high-purity germanium sonde can be used to identify radionuclides buried in old disposal trenches. Detailed records were not kept for many of the early waste disposal sites and little is known about the radioactive sources present. For instance, ^{152}Eu, ^{154}Eu, $^{178\mathrm{N}}$Hf, and ^{101}Rh are not listed as being present at this site in the published references.

More field work is needed under a wider variety of borehole conditions to provide a basis for selecting the optimum detector system or systems for monitoring nuclear waste disposal sites. In addition to calibration test tanks, a computer model might provide a practical foundation for quantitative radioisotope analyses under the wide variety of field conditions that exist at these sites. A prototype variable borehole model has been built and a computer model is under development, but considerable effort will be required before either of the detector systems briefly described in this report can be used for routine monitoring at nuclear waste disposal sites.

ACKNOWLEDGMENTS

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