CONF-9506235--1
UCRL-JC-119236
PREPRINT

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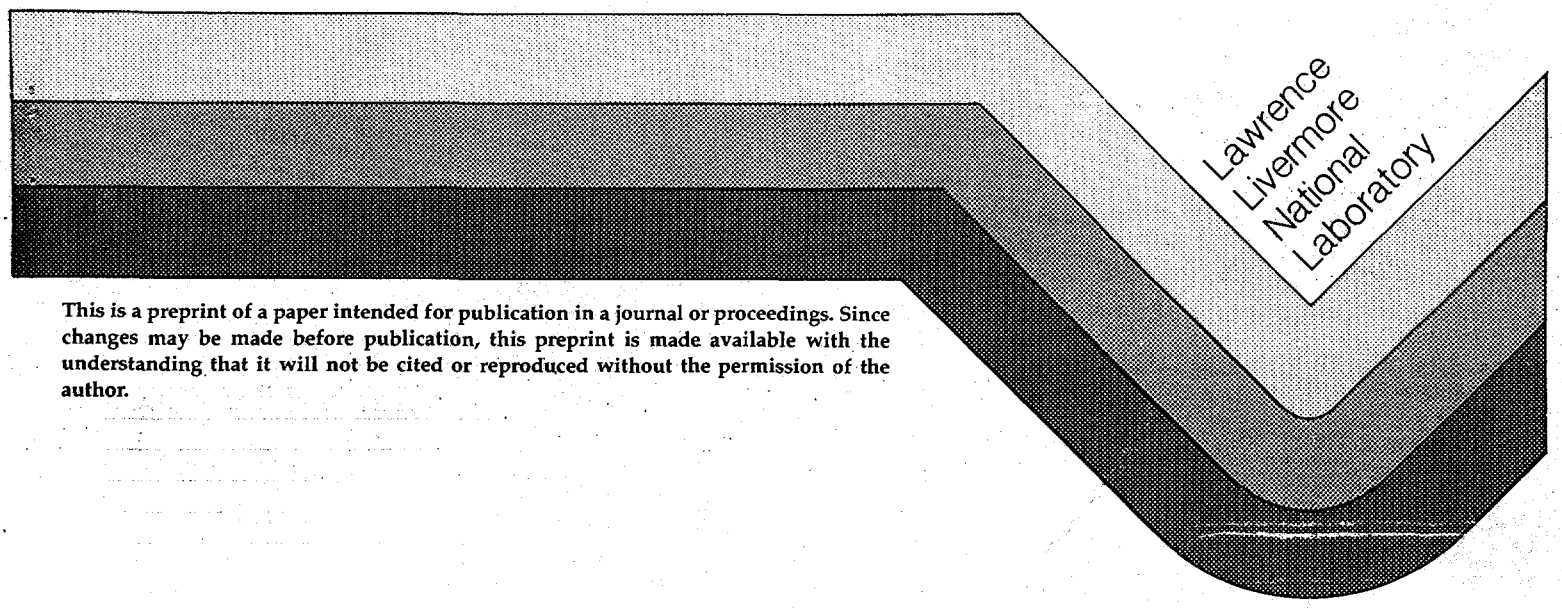
This paper was prepared for submittal to
International Conference on Environmental
Monitors and Hazardous Waste Site Remediation
Munich, FR Germany, June 19-23, 1995

May 26, 1995



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A field strategy to monitor radioactivity associated with investigation derived wastes returned from deep drilling sites

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ABSTRACT

The U.S. Department of Energy, Nevada Operations Office, Underground Test Area Operable Unit (UGTA) is drilling deep (>1500m) monitoring wells that penetrate both unsaturated (vadose) and saturated zones potentially contaminated by sub-surface nuclear weapons testing at the Nevada Test Site, Nye County, Nevada. Drill site radiological monitoring returns data on drilling effluents to make informed management decisions concerning fluid management. Because of rapid turn-around required for on-site monitoring, a representative sample will be analyzed simultaneously for α, β and γ emitters by instrumentation deployed on-site. For the purposes of field survey, accurate and precise data is returned, in many cases, with minimal sample treatment.

A 30% efficient high purity germanium detector and a discriminating liquid scintillation detector are being evaluated for γ and α / β monitoring respectively. Implementation of these detector systems complements a successful on-site tritium monitoring program.

Residual radioactivity associated with underground nuclear tests include tritium, activation products, fission products and actinides. Pulse shape discrimination (PSD) is used in α / β liquid scintillation counting and is a function of the time distribution of photon emission. In particular, we hope to measure ^{241}Am produced from ^{241}Pu by β decay. Because ^{241}Pu is depleted in fissile bomb fuels, maximum PSD resolution will be required. The high purity germanium detector employs a multichannel analyzer to count gamma emitting radionuclides; we will designate specific window configurations to selectively monitor diagnostic fission product radionuclides (i.e., ^{137}Cs).

1.0 INTRODUCTION

Since 1951, the Nevada Test Site (NTS), Nye County, Nevada has been the continental proving ground for United States and British nuclear weapons testing. Since 1955, 828 underground experiments have been fired at the NTS. Approximately one-third have been detonated beneath or within one cavity radius of the water table. As a part of a Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) strategy to address the potential for groundwater contamination associated with underground nuclear testing, the U.S. Department of Energy (DOE), Nevada Operations Office, Environmental Restoration Division designated the Underground Test Area Operable Unit to manage a remedial investigation / feasibility study (RI/FS) of NTS groundwaters impacted by testing. A part of the RI/FS incorporates a dedicated deep (>1500m) drilling program to determine the nature and extent of potential contamination proximal to testing centers. Since 1991, 24 wells have either been drilled or rehabilitated. The first phase of drilling concentrated on uncontaminated site locations downgradient from testing centers where data was returned on hydrogeology and the groundwater flow system. Presently the drilling focus has shifted to targets within the testing centers to better characterize the migration of dissolved radionuclides. The first site for 'near-field' drilling is 275m downgradient from the 1975 TYBO nuclear test which was fired in NTS hole U20y at a depth of 765m in Area 20. The measured TYBO pre-shot static water level was 630m. The drilling strategy calls for completion of a three well cluster with all three wells drilled from the same pad. The first well will be completed to a depth of at least 915m. In order to make informed decisions about the management of investigation derived waste returned from deep drilling, on-site radionuclide monitoring provides field and regulatory personnel a rapid screening technique to evaluate the waste stream in 'real-time'.[1] Besides meeting regulatory requirements, 'real-time' monitoring also affords drilling and field personnel maximum protection against exposure to radionuclides. Fluids circulated through the drill string will be monitored every 9m during drilling. Returned drilling fluids with tritium concentrations more than 200,000 pCi/L, gross α more than 150 pCi/L or gross β more than 500 pCi/L will be contained in lined sumps until the liquid has evaporated. Concentrations below these thresholds may be discharged to unlined infiltration areas.

Radionuclides particularly targeted for identification and early detection by field based monitoring are ^{3}H, ^{60}Co, ^{85}Kr, ^{125}Sb, ^{137}Cs and ^{241}Am. In particular, ^{3}H, ^{85}Kr and ^{125}Sb are produced in forms that are almost completely

mobile in groundwater. These species serve as conservative indicators of radionuclide migration because they are not attenuated by the rock through which they flow.

2.0 TRITIUM MONITORING

Tritium is currently being used as an early indicator of the potential migration of radionuclides in NTS groundwaters. Because it is conservative, tritium will not be depleted nor enriched in groundwaters by chemical processes including ion exchange, speciation, dissolution or precipitation; its presence is an unambiguous indication of subsurface contamination associated with underground nuclear weapons testing. A program to monitor tritium during drilling operations and well development was successfully initiated in 1992 for all UGTA drilling at the NTS. This procedure allows tritium to be monitored at the drill site hourly during continuous three-shift operations.[2]

Three Packard 1600 TR liquid scintillation counters (LSCs) are dedicated for this effort. For 'near-field' drilling two LSCs will be assigned to the drill-site facility and the third used as a backup. To protect the delicate instrumentation during frequent relocation, each counter has been 'hardened' by strapping the lead housing around the detector securing it to the counter frame. In addition, shock mounts were installed between the counter and the cart support in each of the four corners. Power is supplied by on-site generators. Line fluctuations from erratic signals and background interferences require line conditioners and a chargeable uninterrupted power supply. The effluent matrix can be made up of any combination of air, water and foam containing DOE approved polymer drilling mud. Because of particulate loadings in the drilling fluid, high pressure filtration is essential prior to scintillation counting. Special chemiluminesensce subtraction routines have been added to the software for the counting system. Radiological controls technicians (RCTs) are trained to collect, filter, and count samples during 24 hour well drilling and development operations.

3.0 SYSTEMS EVALUATION

3.1 Field deployed gamma detectors

For 'near-field' drilling, stand-alone tritium monitoring is less diagnostic as a comprehensive indicator of contamination. Several fission products including ^{137}Cs and ^{90}Sr have volatile gaseous precursors (^{137}Xe and ^{90}Kr respectively) that may be injected as gases during the detonation and may be heterogeneously redistributed, particularly in the vadose zone, and missed by a stand-alone tritium monitoring program. In addition, while tritium is diagnostic of the presence of radionuclides, the identity of specific fission products can only be confirmed by spectral gamma counting.

Frequently drilling fluid returns incorporate natural ^{232}Th, ^{238}U and ^{40}K both from bentonite-based drilling fluids as well as the evolved tuffs which comprise much of the geology of the Nevada Test Site.[3] The spectral gamma detectors are capable of differentiating between natural and anthropogenic radionuclides in the field.

Historically, high purity germanium gamma detectors have been field deployed. However, early systems were cryogenically cooled with liquid nitrogen, were extremely heavy (90kg) and incorporated a high pressure gas system which requiring venting. Each of these presents substantive logistical problems at the Nevada Test Site. In particular, remote drilling sites may be far removed from convenient source of liquid nitrogen. For this reason, the germanium detectors are cooled using a refrigeration unit charged with an environmentally safe compound. The 20% and 30% efficient Canberra high purity germanium (HPGe) detectors have a resolution of 1.9 eV at 1332 MeV. Component Canberra 2002 preamplifiers and 2020 amplifiers are both low noise. Canberra Accuspec A acquisition interface ADC analyzer boards are designed to run on a DOS PC platform. All analysis and data reduction software has been configured to be easily operated and interpreted by RCTs in the field.

Calibration factors must be developed to convert a measured full-absorption peak count rate to activity in the drilling fluid. The appropriate factors for a given detector are derived using three parameters: the manufacturer's quoted efficiency at 1332 KeV relative to a 3X3 NaI detector, the detector's orientation in the field (up or down) and the Ge crystal length/diameter ratio. The accuracy of the results using this simplified calibration technique is estimated to be 10 to 15%.[4]

The germanium systems have been evaluated using samples of drilling fluid contaminated with short-lived fission products in an attempt to mirror field conditions and determine the sensitivity, efficiency and reproducibility of the fielded systems (Figure 1 - Figure 4). We contrast the performance of both 20% and 30% efficient HPGe detectors.

The following table shows the typical distribution of the short-lived fission products in actual post-event drill-back fluids. The U4u sample was collected in 1994 by a down-hole evacuated bailer lowered into the DALHART post-event drill-back hole at a depth of 533.5m. The U2bs sample was collected in 1973 after the STARWORT event from a depth of 658.5m and recovered from archives for this study. A 400 ml glass jar offers the maximum geometry for the lead shielded counting chamber appropriate for these detectors.

MATRIX DRILLING FLUIDS ANALYZED BY 20% and 30% EFFICIENT GERMANIUM DETECTORS (in pCi per 400ml sample)

U4u (1994) Post Event	Isotope	20% Detector (1σ error)	30% Detector (1σ error)
"	^{60}Co	494 (4.1%)	411 (4.1%)
"	^{125}Sb	82 (18%)	82 (10.7%
"	^{137}Cs	868 (9.3%)	887 (3.6%)

U2bs (1993) Post Event	Isotope	20% Detector (1σ error)	30% Detector (1σ error)
"	^{60}Co	49798 (3.1%)	Not Available
"	^{125}Sb	Non-detect	Not Available
"	^{137}Cs	3225 (4.7%)	Not Available

Limits of detection of ^{241}Am in drilling muds were determined using the 20% and 30% efficient HPGe detectors. A clean reference matrix mud was collected from an uncontaminated well drilling site during 1994. This clean mud was spiked with aliquots of U2bs archived sample (which contained ^{60}Co) and an ^{241}Am source in the proportions tabulated below. The ^{241}Am source was counted in a volume less than 400 ml.

^{241}Am MATRIX SPIKES COMPARED (in pCi/g)

SPIKE #	^{241}Am SPIKE (g)		CALCULATED ACTIVITY		20% DETECTOR OBSERVED ACTIVITY (1σ error)	30% DETECTOR OBSERVED ACTIVITY (1σ error)
Source Activity			270		243 (2.6%)	298 (2.5%)
1	0.1		27		Non-Detect	Non-Detect
2	0.2		54		92 (40%)	Not Available
3	0.6		168		651 (16.2%)	211 (29.1%)
4	1.0		270		520 (18.1%)	Not Available
5	2.5		675		1431 (11.0%)	1708 (10.5%)

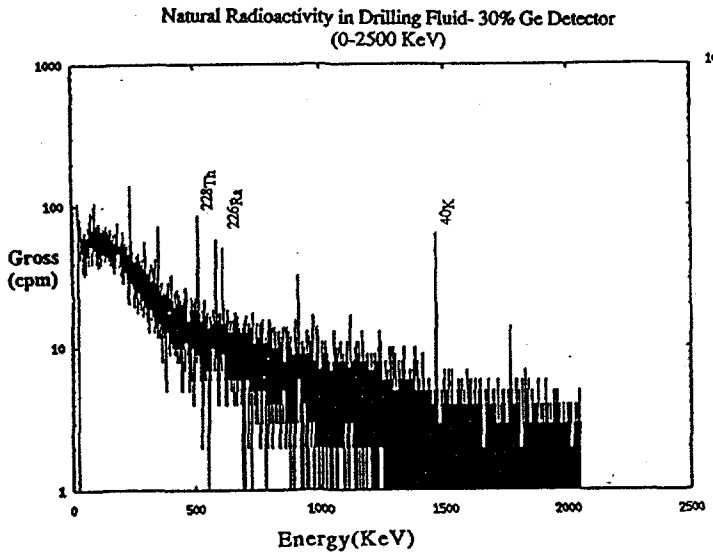

Figure 1.

Matrix drilling fluid. Spectra displays natural background peaks from ^{228}Th, ^{226}Ra and ^{40}K.

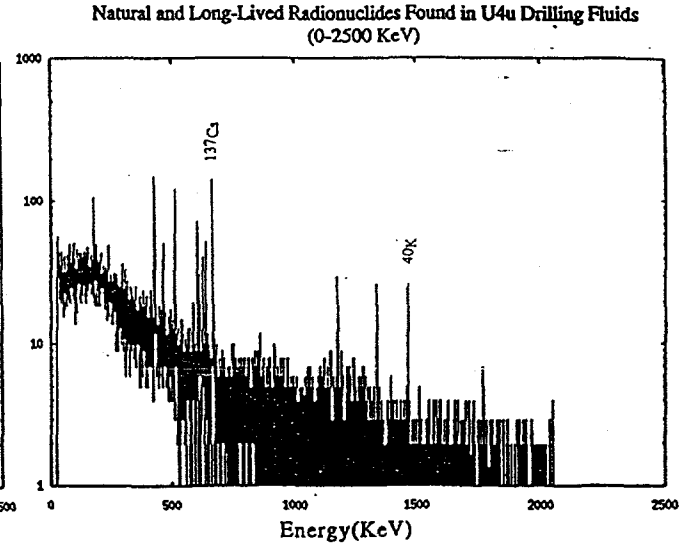

Figure 2.

Drilng fluid containing radionuclide contamination from nuclear weapons testing ^{60}Co, ^{125}Sb, and ^{137}Cs plus the natural background peaks resident in drilling fluid as seen in Figure 1.

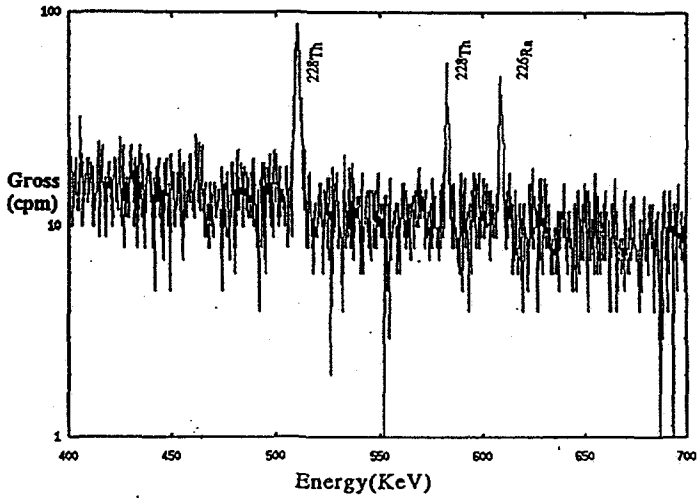

Figure 3.

Spectra is an expanded region from 400 to 700 KeV showing the natural background - Figure 1. The identified peaks are ^{228}Th(511, 583), and ^{226}Ra at 609 KeV.

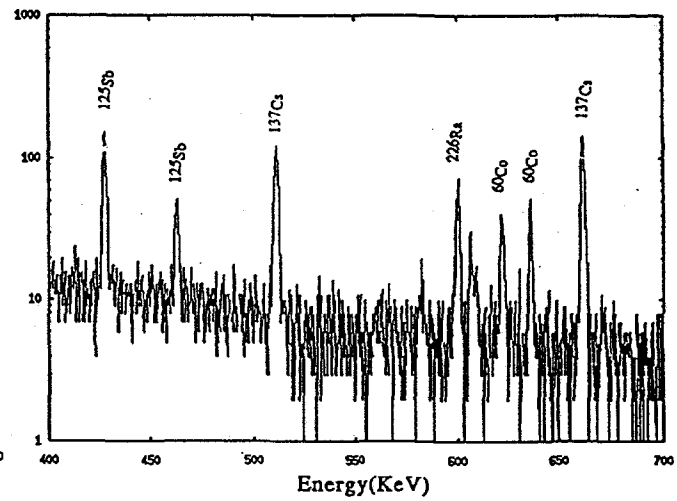

Figure 4.

Spectra displays the expanded region of 400 to 700 KeV - Figure 2. The first peaks are ^{125}Sb (428 and 463 KeV) followed by the natural peaks for ^{228}Th at 511 Kev.

^{60}Co MATRIX SPIKES COMPARED
(in pCi/g)

SPIKE #	U2bs SPIKE (ml)		CALCULATED ACTIVITY		20% DETECTOR OBSERVED ACTIVITY (1σ error)	30% DETECTOR OBSERVED ACTIVITY (1σ error)
			125			
2	2.0		250		295 (11.1%)	Not Available
1	4.0		500		Not Available	480 (9.0%)
3	5.0		625		420 (7.3%)	671 (6.2%)
4	8.0		1000		768 (5.9%)	Not Available
5	10.0		1250		1099 (4.8%)	1854 (3.4%)

The drill-site protocol calls for a 400 ml glass jar to be filled with the drilling fluid without further sample preparation. The 20% efficiency Ge detector will serve to prescreen each hourly collected sample. The 20% sample will be transferred to the 30% efficient Ge detector for further counting as a new sample arrives the next hour for prescreening.

3.2 Alpha / beta discrimination

The discriminating liquid scintillation detector for α / β monitoring exhibits excellent resolution for ^{241}Am with a characteristic 5.27 MeV α emission. The selection of ^{241}Am as a target isotope is significant for two reasons: 1) diagnostic alpha and gamma emission energy peaks and 2) ^{241}Am is produced by β decay of ^{241}Pu with a half-life of 14.4 years. Weapons grade plutonium consists of approximately 0.5% ^{241}Pu which persists after detonation as unburned nuclear fuel. However, because ^{241}Pu and ^{241}Am are depleted in weapons fuels and are refractory species which are relatively insoluble in groundwater their presence is not expected in 'near-field' drilling returns.

The energy range for most beta emitters is 0-2 MeV; and for alpha emitters is 3-8 KeV. The peaks are shaped very differently. Pulse shape discrimination allows mixed α and β emitters in a single sample to be resolved by the time distribution of the light emission each generates from the scintillator. LSC/PSD techniques allow the measurement of alpha particles with excellent spectral resolution and low background. However counting efficiency by this technique is low and sample preparation can be complex.

Results for α / β discrimination using a Beckman 6500 that has both quench correction and chemiluminesence software are provided below. The ^{241}Am standard activity is 270 pCi/g. Results are also listed for ^{241}Am mixed with a ^{36}Cl standard and ^{241}Am mixed with a ^{3}H standard. The table clearly shows the ability of the α / β discriminating LSC to resolve α emitters in α - β mixtures relative to β screening alone.

EVALUATION OF α /β DISCRIMINATION TO DETECT ^{241}Am IN α - β MIXTURES
(in pCi/g)

NUCLIDE	β PROTOCOL (1σ error)	α / β PROTOCOL (1σ error)
^{241}Am	277 (1.0%)	210 (1.2%)
^{241}Am + ^{36}Cl	1411 (0.05%)	161 (1.3%)
^{241}Am + ^{3}H	1295 (0.05%)	232 (1.1%)

4.0 RESULTS AND FIELD PROTOCOL

During 'near-field' drilling short count tritium scans will be processed every quarter hour by the LSC procedure established in 1992. For unsaturated and saturated zone drilling in the 'near-field', tritium monitoring will be augmented by drill-site gamma monitoring.

The analysis of drilling fluid effluents using a dedicated 30% efficient HPGe detector enhances UGTA on-site radionuclide contaminant diagnostic capability. The capability to correctly and quickly identify single fission

products and distinguish natural from contaminant radioactivity provides health and safety personnel necessary information to adequately manage the investigation derived waste stream while minimizing interruptions to an expensive and time-critical drilling schedule.

As a result of our work, we significantly improved the limit of detection of ^{241}Am using LSC discrimination. The gamma ray peak for ^{241}Am occurs at 59.5 KeV at the height of the background continuum. By comparison, the 5.27 MeV α particle is easily resolved by the α / β LSC.

5.0 NEW DEVELOPMENTS

Investigators at the Lawrence Livermore National Laboratory (LLNL) have developed a compact, rugged, and high-resolution electro-mechanically cooled high-purity germanium detector for remote / field applications. This system offers lightweight portability and low power consumption. Superior energy resolution is achieved using the break-through active vibration suppression technology which is comparable to (and in many cases better than) conventional liquid nitrogen cooled HPGe systems. The system weighs 6.8 kg and requires 40 watts of power to operate once the detector is cooled to its operating temperature.[5] The detector has been developed to provide safeguard measurements of special nuclear materials indicative of isotopic composition and enrichment signatures; the system has been successfully field tested using plutonium and uranium metals. This technology is predicted to extend the life expectancy of the detector about a factor of 10. This LLNL detector can be operated on a 18 to 32 volt battery for a period of about 8-12 hours.[5] Advances in component miniaturization promise extremely portable gamma detectors in the very near future with design lifetimes in excess of 10 years. Ultimately, such a detector might be fielded down-hole in the saturated zone to provide in-situ gamma monitoring.

6.0 ACKNOWLEDGMENTS

Gamma detectors used in this investigation were purchased by the IT Corporation for the U.S. Department of Energy and were provided to LLNL for developmental purposes. Field monitoring strategies have benefited from conversations with Linda Cardenas of the IT Corporation, Las Vegas, Nevada. Kurt Schmidt of IT Corporation, Las Vegas helped with gamma detector logistics. Efficiency calibrations for the gamma detectors were computed by Ken Raschke of LLNL.

Work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

7.0 REFERENCES

1. Murphy, E.M., D.D. Hostetler and J.C. Evans "Field testing of environmental sensors: applications of voltametry and fiber optics to ground-water monitoring", *Unpublished Report*, Pacific Northwest Laboratory, Richland, WA, 1995.
2. Rego, J.H. and D.K. Smith, "The development of a versatile field program for measuring tritium in "real-time", *Journal of Radioanalytical and Nuclear Chemistry, Articles*, v. 194, no. 2, pp. 389-392, 1995.
3. Esser, B.K., "The effect of bentonite-based drilling fluids on radiochemical sampling of Nevada Test Site groundwaters", *Position Paper*, Nuclear Chemistry Division, Lawrence Livermore National Laboratory to the U.S. Department of Energy, Nevada Operations Office, 18p., March, 1994.
4. Helfer, I.K. and K.M. Miller, "Calibration factors for germanium detectors used for field spectrometry", *Draft*, Environmental Measurements Laboratory, U.S. Department of Energy, New York, NY.
5. Neufeld, K.W. and W.D. Ruhter, "Portable electro-mechanically cooled high-resolution germanium detector", *Unpublished Report*, Isotope Sciences Division, Lawrence Livermore National Laboratory, 1994.