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Field Test of a Post-Closure Radiation Monitor

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Abstract

The DOE is conducting remedial actions at many sites contaminated with radioactive materials. After closure of these sites, long-term subsurface monitoring is typically required by law. This monitoring is generally labor intensive and expensive using conventional sampling and analysis techniques.

The U.S. Department of Energy's Morgantown Energy Technology Center (METC) has contracted with Babcock and Wilcox to develop a Long-Term Post-Closure Radiation Monitoring System (LPRMS) to reduce these monitoring costs. The system designed in Phase I of this development program monitors gamma radiation using a subsurface cesium iodide scintillator coupled to above-ground detection electronics using optical waveguide. The radiation probe can be installed to depths up to 50 meters using cone penetrometer techniques, and requires no downhole electrical power. Multiplexing, data logging and analysis are performed at a central location.

A prototype LPRMS probe was built, and B&W and FERMCO field tested this monitoring probe at the Fernald Environmental Management Project in the fall of 1994 with funding from the DOE's Office of Technology Development (EM-50) through METC. The system was used to measure soil and water with known uranium contamination levels, both in drums and in situ at depths up to 3 meters. For comparison purposes, measurements were also performed using a more conventional survey probe with a sodium iodide scintillator directly butt-coupled to detection electronics.

This paper presents a description and the results of the field tests. The results were used to characterize the lower detection limits,

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precision and bias of the system, which allowed the DOE to judge the monitoring system's ability to meet its long-term post-closure radiation monitoring needs. Based on the test results, the monitoring system has been redesigned for fabrication and testing in a potential Phase III of this program. If the DOE feels that this system can meet its needs and chooses to continue into Phase III of this program, this redesigned full scale prototype system will be built and tested for a period of approximately a year. Such a system can be used at a variety of radioactively contaminated sites.

1. LPRMS Probe

Because it is intended for installation by a CPT truck, the mechanical design of the Long-Term Post-Closure Radiation Monitor (LPRMS) probe which was fabricated and tested in this program was based on the dimensional envelope of a 10 cm^2 cone penetrometer tool with a 1-7/16 inch (3.65 cm) outside diameter and a conventional 60 degree cone tip angle. The LPRMS probe consisted of a scintillation head housing the scintillator, a detection head housing the PMT and detection electronics, and several 1 meter long threaded extension sections for the push rods and lightguide. The scintillation head incorporated a 2.5 cm diameter by 25 cm long CsI(Tl) scintillator inside a 0.36 cm thick 4130 steel window section which extended slightly past the scintillator on both ends. In this design, the window material carried the push forces applied to the tool; this limited the maximum push force for this tool to about 20 tons. Although it was made in 1 meter sections for CPT installation, for these tests the probe was fully assembled above ground prior to installation and testing.

A drawing of the LPRMS probe is shown in Figure 1. The optical photons from the scintillator were transmitted by a single air-clad PMMA rod 2.5 cm OD. The optical waveguide was directly butt-coupled to the scintillator. To accommodate the waveguide, the bore of the extension sections was increased to 2.7 cm from the normal CPT rod bore of 1.6 cm. The extension sections were 1 meter in length and extended to the surface. At the surface end of the probe, the extension sections were coupled to a detection head containing a 1-1/8" head-on bialkali PMT, a voltage divider base, a pre-amp and pulse shaping electronics. The optical waveguide was directly butt-coupled to the PMT face. The PMT was operated in the pulse mode with a cathode ground (positive high voltage). This mode of operation is consistent with either photon counting or spectroscopic analysis techniques. The PMT was magnetically shielded, and operated at ambient temperature. A relatively thick stainless steel housing was used to minimize short term temperature changes of the PMT.

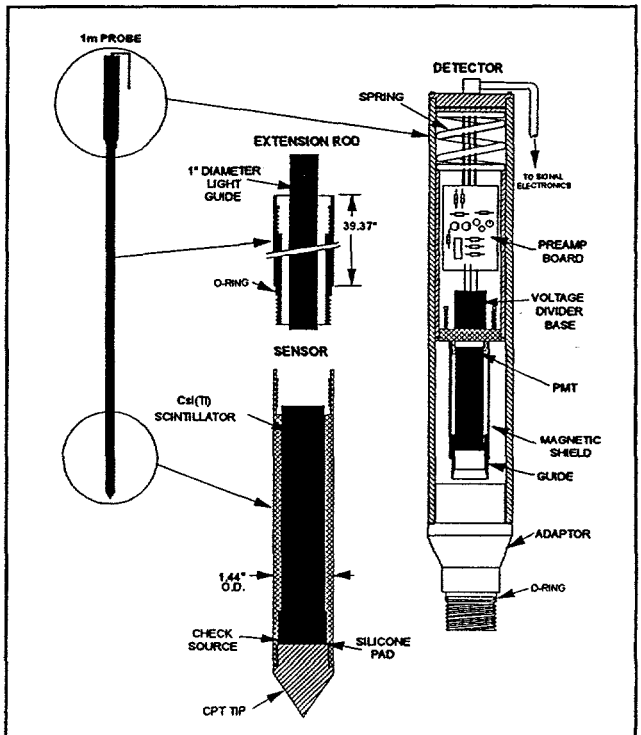

Figure 1. LPRMS Probe

2. Survey Probe

To provide performance comparison data, tests were also performed using a gamma radiation survey probe developed by B&W for radiation survey applications during site characterization and remediation. The survey probe is 1.42 inches (3.6 cm) diameter by approximately 16 inches (40 cm) long. It is designed to be lowered into a 1.5 inch diameter or larger casing on a wireline (logging mode) by hand or from a light tripod using a small hand winch. The probe contains a 1 inch (2.54 cm) diameter by 6 inch (15.3 cm) NaI(Tl) scintillator directly butt-coupled to a bialkali PMT with optical grease. The probe also contains magnetic shielding for the PMT, the voltage divider and a Cockroft-Walton high voltage power supply within a potted housing for moisture and shock resistance. A drawing of this probe is shown in Figure 2.

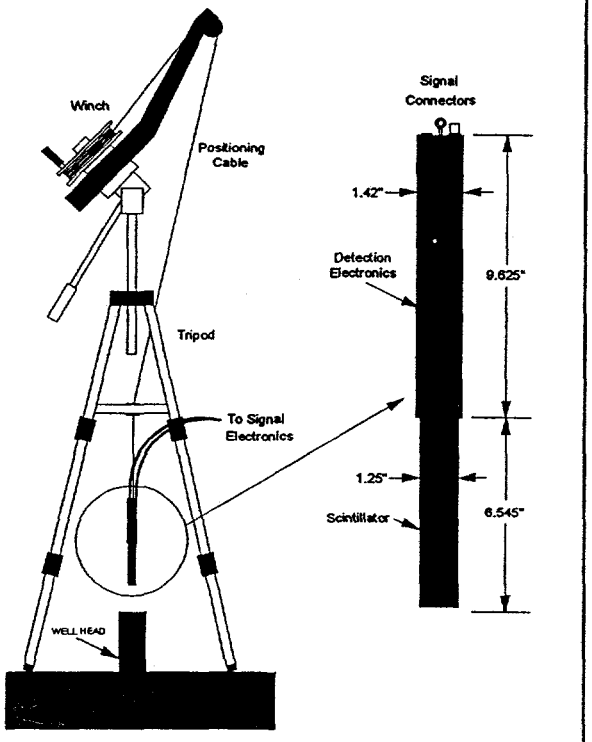

Figure 2. Survey Probe.

3. Gamma Spectrometer

The data from the two probes was acquired, analyzed, stored and printed using a commercially available PC-based two channel gamma spectrometer system. The system, manufactured by Canberra Nuclear, is comprised of two acquisition interface boards (NaI+) installed in a Gateway 486-66 PC plus gamma spectroscopy software (GENIE-PC) to provide the functions of a hardware-based MCA (multi-channel analyzer). The interface boards provide a pre-amp DC power supply, an integrated HV power supply, data amplifier and a 100 MHz Wilkinson ADC (analog to digital converter).

The functions and settings of the interface board hardware are controlled from the software through a window-style graphical user interface. The software operates under an OS-2 operating system and is a true multitasking architecture; the system can thus support simultaneous and fully independent counting and analysis procedures on the two channels. In addition to hardware control, MCA control and basic gamma spectroscopy functions (such as continuum correction and peak searches), the software also performs energy and efficiency calibrations, background subtractions, nuclide identification (interference corrected), spectrum scaling or gain stabilization, calculates weighted mean activity for the nuclides detected and the MDA (minimum detectable activity) for any specified nuclide which is not located in the spectral data. The data from a count procedure is stored in a single extensible file (a Configuration Access Method or "CAM" file) which contains the spectral data, calibration information, analysis parameters, intermediate and final analysis results, setup parameters and the complete analysis library used. The selected results, including the energy spectrum if desired, of the analysis are then output in a

user specified report format to a printer (Hewlett Packard Laserjet).

4. Calibration

To make quantitative measurements, a gamma probe needs both an energy calibration and an efficiency calibration. In normal practice, a source with known isotopic content and activity, and the same geometry as the planned measurement geometry is positioned at the detector. The source is then counted for a fixed length of time. Because the isotopic content is known, the known energy lines can be used to perform the energy calibration. Because the activity levels are known and the geometry is the same as that to be measured, the efficiency calibration can also be readily performed.

For the demonstration tests, the probe will be used inside casings, completely surrounded by contaminated soil. Soil 30 cm or more away from the casing still contributes to the measured signal, as does soil above and below the probe. To duplicate the measurement geometry, the calibration source would need to be roughly the size of a 55 gallon drum (about 2 feet in diameter and about 3 feet high). This is not an available or practical calibration source geometry. Instead, the energy calibration was performed using an Amersham QCD.1 nine nuclide disc source, positioned at the center of the scintillator, side-on. This source provides 11 known energy lines which can easily be used to perform the energy calibration in the lab or field. It is not suitable for the efficiency calibration.

The detector efficiency as a function of energy was determined by calculation in several steps. A spreadsheet model of the soil, soil moisture, casing, detector can and scintillator absorption was used to determine the gammas absorbed within the scintillator volume for known uniform activity levels in the soil. The model also accounted for the scintillation efficiency of the scintillator, optical losses due to reflection and refraction in the scintillator and lightguide, and the PMT quantum efficiency to predict the count rate at the PMT anode for a given soil activity level. This provided a first approximation of the overall efficiency in the soil measurement geometry; this approximation was then adjusted empirically based on counts of a hollow cylindrical source containing known activities of potassium, uranium and thorium. Tables of the efficiency vs energy were then stored in a computer file in the gamma spectrometer computer for use in later analysis; these efficiency values were used for the preliminary analysis of the field data.

Some of the tested soils in the field test (described below) had contamination levels that were reasonably well known, at least for the uranium isotopes. Data from these tests were analyzed and the predicted activities compared to the known activities. The analysis results consistently showed higher activity levels than the laboratory analyses indicating that the calculated efficiencies had been over-corrected by about 20%. The correction factors were then revised based on the field test data and the resulting efficiencies were used for all analyses.

The energy and efficiency calibrations permit the system to identify nuclides and calculate their activities based on a library of gamma lines and yields. To calculate nuclide activities in terms of pCi/gram, the analysis quantity in grams must be known. To determine the sample quantities for analysis, we calculated an effective radius (30 cm) and effective view angles (+- 30 deg) for the probe beyond which the contribution of additional sample material is minimal. Based on these effective dimensions, we calculated the active

sample volume for the probe in cubic centimeters. This volume is multiplied by the sample density to determine the analysis quantity.

5. Test Description

The Fernald Environmental Management Project (FEMP) site at Fernald, Ohio was the selected test site. This site is a U.S. DOE site in southwestern Ohio, approximately 17 miles from Cincinnati. Uranium isotopes are the primary contaminants of concern at this site, resulting from about 35 years of processing of uranium ore concentrates into high purity uranium metal. This test program was coordinated with three other programs: the DOE Uranium in Soils Integrated Demonstration (USID), the DOE Cone Penetrometry Demonstration (CPD) and the B&W funded Survey Tool program. The USID program provided previously characterized soils to be used in fabricating test drums with known activity levels. As part of the CPD program, two locations at the FEMP were sampled and analyzed for uranium contamination vs depth; the bores at these locations were then cased with 1.5 inch PVC casing for later measurement in our program. The B&W Survey Tool program provided the survey probe which was used to generate comparison data for each of the tests performed. All data from both probes was acquired with B&W's laboratory 2 channel gamma spectrometer.

A total of four weeks of testing were performed at the FEMP in the fall of 1994. Two types of tests were performed: tests using drummed samples with known contamination levels and in-situ (sub-surface) tests in cased boreholes at three locations at depths up to four meters. The drummed sample tests included the following types of samples:

- Homogenized soils from the USID program: eight samples with predominantly uranium contamination at known activities from 50 to about 1750 pCi/gram (three duplicates), plus one sample of clean water which was percolated into and retained in one of the samples of contaminated soil for testing;

- Water: three samples with predominantly uranium contamination at known activities from the South plume pumping station, from the storm water retention basin and from the biodenitrification facility;

- Sand matrix/water: one sample of sand matrix at background, plus one sample of water at a known activity level, which was percolated into and retained in the sand/gravel matrix for testing.

The in-situ tests were performed at three locations; one in an existing monitoring well, and two in boreholes available from the Cone Penetrometer Demonstration (CPD) test program which were subsequently cased with PVC. Over 200 counts were performed with each probe, with count times varying from 3 to 90 minutes. The system performance results presented in this paper were determined based on the counts and analyses of the drummed soil samples.

The test specimens for the drummed soils tests consisted of eight 55 gallon drums of characterized soils from the USID with five different activity levels; the nominal activity levels of the soils are listed in the table below. For each of the first 4 test specimens listed above, sufficient soil to fill the drums was taken from larger boxes of soil which had previously been sampled and analyzed for uranium isotopes. These laboratory analysis results have

Soils Samples for Drummed Soils Tests

Sample	Drum ID	Activity (pCi/gram)	Test ID
CP	F-392	51	1B
C-35	C-389	95 (two drums)	1D & 1F
C-100	D-389	146 (two drums)	1C & 1G
C-200	E-388	311 (two drums)	1E & 1H
---	P011-0380	> 1000	2A & 2B

been used for the comparisons contained in this paper; no analyses of the drummed soils were performed. The analysis results showed considerable spread over the sampling locations within the box. The analysis results thus provide only a general indication of the isotopic uranium activity of the drummed soils, not their actual content. Sample P011-0380 was taken from a similar box, but was characterized only with a single grab sample (total U greater than 1000 pCi/g); data from this sample were thus not used for the system performance.

6. Performance Results

6.1. Lower Detection Limits

The normal procedure for determining the lower detection limits (LDL) by isotope is to count and analyze the Minimum Detectable Activity (MDA) for a "blank", a sample identical to the unknowns in geometry, background nuclides (such as K-40) and absorption characteristics, but with no other isotopic activity. The count protocols and analysis parameters used are identical to those used to count and analyze unknowns. A blank soil sample was not available for the tests performed at FEMP. However, one of the test runs provided a reasonably close match to a blank: run 3A, a drum of clean sand with K-40 activity of 6.4 pCi/g.

For this sample, both the LPRMS probe and the B&W Survey Probe had been used to perform 30, 60 and 90 minute counts. The LPRMS probe was in the 1 meter configuration for test 3A. A Genie-PC nuclide library was prepared which included all of the gamma emitting isotopes from the list, prepared in Phase I, of nuclides found on DOE lands. This library included short half-life daughters which could reasonably be expected to be in secular equilibrium with the parent, with yields and half lives adjusted to provide the MDA of the parent, based on detection of the daughter. An MDA analysis was performed for both of the probes for test 3A using this library. This analysis was performed using Genie-PC, which uses the method of Currie for MDA calculation, at 95% confidence. The table below shows the MDA values for uranium isotopes from this analysis, for 30 minute and 90 minute count times. The isotopic MDA is defined as the lowest line MDA for any of the isotope's gamma lines.

This table shows that the LDLs for the LPRMS probe are generally about twice those of the survey probe, except for isotopes which only have low energy gamma lines. Both probes show LDLs for U-235, U-237 and U-238 which are potentially useful for monitoring applications; the ratio of the 30 and 90 minute count LDLs shows that the LDLs are dominated by count statistics, and that longer count times could be expected to further reduce the LDL. For reliable measurement, it's

Table 1. Lower Detection Limits: Uranium
30 and 90 Minute Counts: Test 3A

Isotope	Survey Probe		LPRMS Probe (1 m)	
	30 min	90 min	30 min	90 min
U-233	130.8	75.5 pCi/g	219.1	126.7 pCi/g
U-234	207.8	119.6 pCi/g	23320	13472 pCi/g
U-235	0.39	0.23 pCi/g	0.52	0.30 pCi/g
U-236	207.5	69.4 pCi/g	2436	1408 pCi/g
U-237	0.68	0.39 pCi/g	1.27	0.74 pCi/g
U-238	4.38	2.53 pCi/g	10.5	6.10 pCi/g

desirable for the activity to be roughly a factor of 5 to 10 or more above the lower detection limit. With the isotopic ratios typical for FEMP, this corresponds to about 50 pCi/g total U for the survey probe and about 125 pCi/g total U for the LPRMS probe for 90 minute count times (based on U-238). For 30 minute count times, the LDLs correspond to about 90 pCi/g total U for the Survey Probe and about 200 pCi/g total U for the LPRMS probe, based on U-238.

6.2. Precision and Bias

Because of the limited number of test articles and their generally low uranium activity levels, the precision values for the LPRMS and Survey probes are stated in Table 2 at isotopic activities of about 5 times the MDA, or roughly 6 to 10 times the LDL, rather than at the more typical 10 times MDA. The values listed in the table are for single 30 minute counts rather than an average of multiple counts. The activities listed in the table are isotopic activities. The precision values given are relative uncertainties; to calculate these values, the measurement uncertainty (at 1 standard deviation) for an activity determination is divided by the activity, and multiplied by 100 to give a percentage (relative precision). All of the measurement uncertainties were calculated using Genie-PC, as part of the analysis sequence.

To determine the bias of the activity measurements, the difference between the measured and known activities was divided by the known activity and the result is multiplied by 100 to give bias as a percentage (relative bias). For the tests at FEMP, the activities in the test drums were only approximately known. The bias values shown in the table below were calculated using the average isotopic activities for the boxes of USID soils as the "known" value, although there will be some unknown bias due to the sampling involved with removing the soils from the boxes and placing them in the drums, and due to the unknown uncertainties of the reference analyses themselves. The bias values were calculated for the same 30 minute counts used in the determination of precision, above.

Table 2. Precision for Detected Uranium Isotopes
Isotopic Activities at about 5 x MDA

	Survey Probe		LPRMS Probe (1 m)	
Isotope	Precision	Activity	Precision	Activity
U-235	5.6%	3.451 pCi/g	7.3%	6.693 pCi/g
U-238(Th-234)	25.0%	21.80 pCi/g	--	n/d
U-238(Pa-234m)	4.7%	155.6 pCi/g	7.4%	155.6 pCi/g

Table 3. Bias for Uranium Isotopes
Isotopic Activities at about 5 x MDA

	Survey Probe		LPRMS Probe (1 m)	
Isotope	Bias	Activity	Bias	Activity
U-235	+3.2%	3.451 pCi/g	+78.2%	6.69 pCi/g
U-238(Th-234)	+382%	21.80 pCi/g	--	n/d
U-238(Pa-234m)	+8.9%	155.6 pCi/g	+5.2%	155.6 pCi/g

7. Discussion

In general, for any given test configuration, it was more difficult for the analysis software to locate peaks for the LPRMS count data than for the survey probe. When the peaks were located in the spectra, more difficulty was encountered in identifying nuclides. For identified nuclides, the uncertainties in the calculated activities were larger. This was due primarily to the poorer resolution of the LPRMS, even though the count rates with the LPRMS probe were 10 to 40 percent higher than with the survey probe.

The effect of resolution on the energy spectrum is illustrated in Figure 3, which shows counts of a calibration source performed with the LPRMS and survey probes. The cal source and count times were the same for both probes. The resolution of the LPRMS probe is about 11.8%, the survey probe resolution is about 7.3% (at 662 keV). The peaks for the LPRMS probe (dotted line) are lower and broader than those for the survey probe (solid line), although they contain about the same or greater number of counts. The net height of the Cs-137 peak at 662 keV is roughly 7500 counts for the LPRMS probe and 12000 counts for the survey probe. The signal-to-noise ratio (net peak height/continuum) for this peak is about 4 for the survey probe and less than 2 for the LPRMS probe. This results in a significant increase in the minimum detectable activity: small peaks, either from low activities or from low yield isotopes, can't be separated from the statistical variation of the continuum count rate.

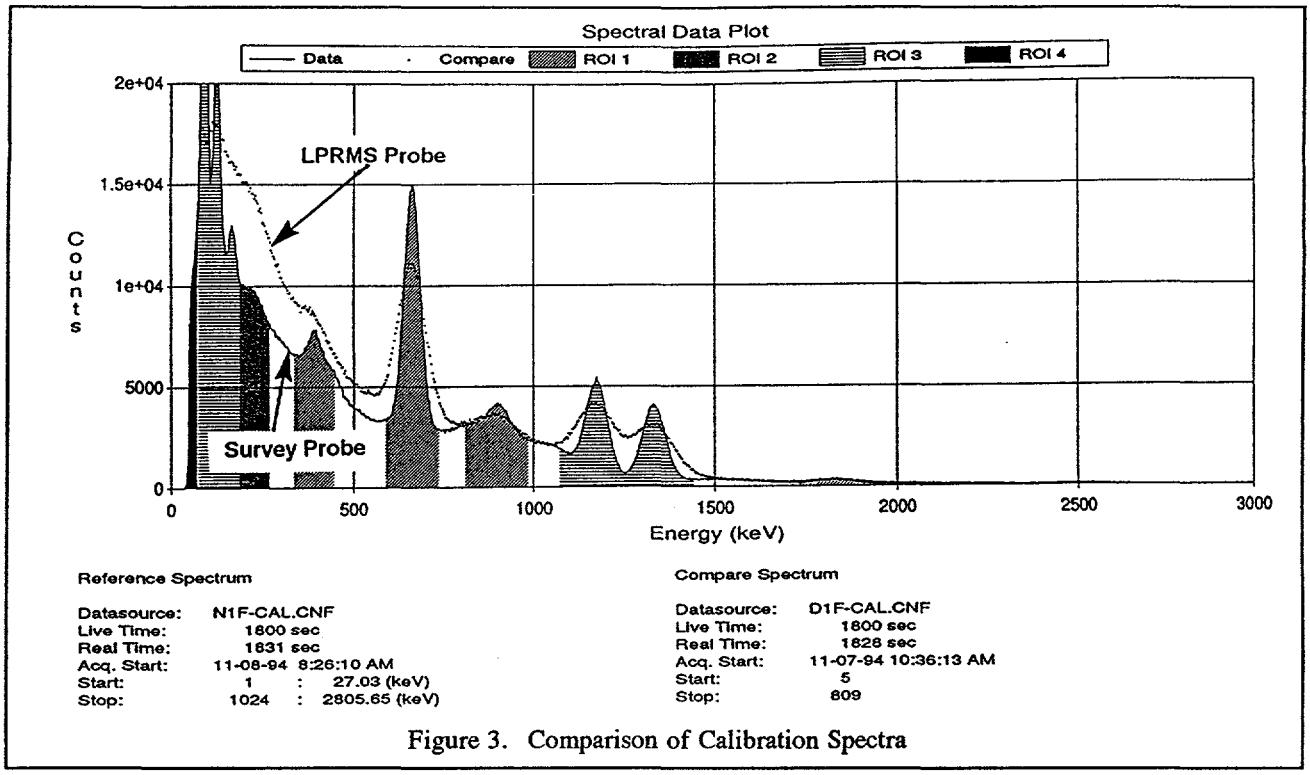

Figure 3. Comparison of Calibration Spectra

The higher FWHM results in greater uncertainty in the location of the peak centroids, making peak identification more difficult. It also results in complications in separating multiple peaks and in determining their areas. For example, the Co-60 peaks in Figure 3 are at 1173 and 1333 keV, separated by 160 keV. With the survey probe, these peaks are cleanly resolved with little overlap, and the Compton edge of the 1333 keV peak (at 1119 keV) is below the ROI for the 1173 peak. With the LPRMS probe, these two peaks have significant overlap, and the Compton edge of the 1333 keV peak is within the 1173 keV peak. While these two peaks can still be separated, simple peak height analysis algorithms will have difficulty correctly determining the peak areas because of the relatively shallow valley between them and the presence of the Compton edge of one peak within the area of the other. More complex analysis routines using interactive Gaussian fits could do a better job of analyzing these peaks, but are more expensive, slower, and can require a priori knowledge of peak locations to be effective.

The controlling factor to the achievable resolution in a PMT/scintillator combination is statistical broadening, based on the number of photoelectrons emitted from the PMT photocathode. This quantity is controlled by the number of incident optical photons/gamma event and the quantum efficiency of the photocathode. In the LPRMS, both of these factors are important. The optical losses associated with using an optical waveguide reduce the number of optical photons incident at the photocathode by about 9 dB, compared to a butt-coupled geometry. The spectral mismatch between the CsI(Tl) emission spectrum and the photocathode response spectrum introduces an additional loss of about 3.5 dB, compared to a bialkali PMT and NaI(Tl) scintillator. All other losses, such as gamma attenuation by the steel scintillator window, are minor by comparison.

The losses associated with the waveguide are primarily due to its limited view angle into the scintillator, a function of its numerical aperture. The waveguide chosen has an NA of about 0.65. Significantly increasing the lightguide numerical aperture to increase the view angle is not practical, because transparent materials with the required higher index of refraction are not readily available. The CsI(Tl) scintillator emission spectrum minimizes the throughput losses in the lightguide; changing the scintillator to improve the spectral match to a PMT would result in a greater increase in the throughput losses than could be gained in a better spectral match. Changing the waveguide to a material with lower losses in the emission spectrum of NaI(Tl) would result in a lower NA and consequently greater view angle losses than would be gained by the decrease in spectral losses.

8. Field Test Conclusions

The critical performance parameters for a post-closure monitor are the lower detection limits and precision. The performance of the LPRMS probe was consistently poorer than that of the survey probe in both of these areas. With 30 minute count times, neither of the probes tested clearly demonstrated the capability of identifying and quantifying uranium isotopes at activities near the post-closure concern levels assumed for this program (35 pCi/g total U, 17 pCi/g U- 238, 0.85 pCi/g U-235). With 90 minute or greater count times, the performance of the survey probe has marginally adequate lower detection limits for both U-235 and U-238, with precision of about 5%. To detect and monitor these isotopes at such activities, the waveguide-coupled LPRMS will require significant improvements in resolution, peak-to-total ratio, or both. A review of the options for reducing optical losses and improving the performance of this design showed that it is unlikely that the needed improvements can be obtained.

Based on the results obtained with the survey probe, it is believed that a resolution of 7.5 to 8.0% (at 662 keV) will be adequate, with some improvement in peak-to-total ratio, to reliably monitor U-235 and U-238 at the assumed post-closure concern levels, using 2 hour count times. We considered the available options to accomplish this, and concluded that a workable approach is readily available, employing a butt-coupled scintillator/PMT probe. This previously rejected approach is now practical for post-closure monitoring, because of the recent development of CPT technology to push low cost plastic casing to depths comparable to those attainable with CPT tools, opening the option for readily retrievable downhole electronic components. This approach retains the desired benefits of low installed cost, serviceability, CPT installation and minimal potential for cross-contamination both during installation and in service. A prototype system based on this approach has been proposed to DOE for demonstration in Phase III of this program; the architecture for this system is shown in Figure 4 below.

9. Cost Comparison

Based on the cost estimates obtained for the Phase III prototype system, the system installed costs have been estimated. These costs do not include any reduction in cost for quantity, marketplace competition or increased maturity of the system technology, and thus can be considered as typical of the first installed system. Costs are included for hardware, software and quality assurance, as well as project management costs for deploying the system. Costs are not included for site specific activities such as determination of monitoring

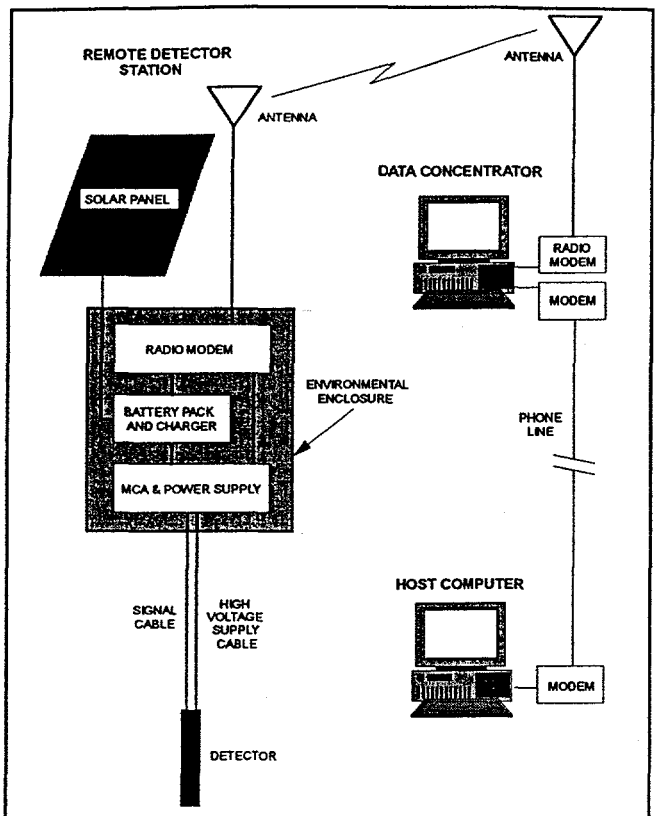

Figure 4. Phase III System Architecture

locations and depths, project specific health and safety plans, permitting and other similar activities.

The system components include those which are required for each monitoring location (PVC casing, completion, probes and remote stations), those which are required for each site monitored (transceiver and data concentrator) and those which are required for the monitoring system as a whole (host computer). For comparison purposes, a system with 12 monitoring locations at 20 meter depth has been assumed, with a maximum distance of 15 kilometers between the remote stations and the data concentrator. This system requires a single data concentrator and a single host computer. It has been assumed that the host computer is dedicated to this one system, although in actuality, a single host can provide analysis and trending for multiple sites. The total system cost for a single system to monitor 12 points with a dedicated host computer is shown below.

Table 4. Installed Costs:Monitoring System

Per Point Costs:	12 @ $20,955	= $241,860
Per Site Costs:	1 @ $38,470	= $ 38,470
Per System Cost:	1 @ $44,085	= $ 44,085
	Total System Cost	= $324,415

For comparison, costs were obtained from FERMCO and Rocky Flats for conventional sampling and laboratory analysis. The total costs per sample were estimated to be about $3500 per sample, with a turnaround time of 60 to 400 days. This estimate does not include costs for project specific health and safety plans, oversight personnel, radiological control technicians, sample shipping or surveying. The costs of conventional sampling and analysis were evaluated using a simple annuity calculation (present worth) assuming an interest rate of 5%. The effects of inflation were ignored. The present worth of the cost of conventional sampling and analysis for 12 locations for 25 years is shown in Table 5 below for analysis intervals of once per year, and once per quarter. With a sampling interval of once per year, the savings with the LPRMS system are about 45%; with an interval of once per quarter, the savings are about 87%.

Table 5. Present Worth of Conventional Sampling and Analysis Costs

Once/year:	$ 591,948
Once/quarter:	$2,367,792

monitoring without significant cost impact, and eliminates the long lead time encountered with conventional sampling and laboratory analyses. Sampling error is eliminated, and each measurement is taken in the same physical location, providing an improved ability to track small changes in activity with good precision. The potential for smearing and cross contamination from sampling operations is also eliminated, as are cutting disposal and grouting. The potential for worker exposure during sampling and sample handling is also eliminated. Use of this system thus provides a faster, better, cheaper and safer means to perform long-term *in situ* monitoring for radionuclide contamination.

10. Acknowledgements

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