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7. Abstract <i>11/2 174/11/20/11</i> High resolution passive gamma-ray logging, high resolution gamma-ray-emitting nuclides in areas surrounding underground waste disposal facilities on the U.S. Department of Energy's Hanford Site. Gamma-ray source concentrations are derived from log data by calculations that employ the calibration factors and correction functions described in this report. Calibration data were collected with a Radionuclide Logging System. Analyses of the calibration data established: (1) calibration factors for potassium, uranium, and thorium, and (2) a calibration function that permits assessments of cesium-137, cobalt-60, and other artificial nuclides not represented in the calibration models.		
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CALIBRATION OF THE RADIONUCLIDE LOGGING SYSTEM GERMANIUM DETECTOR

Russel Randall
August 16, 1994

Introduction

Calibration factors have been determined for the high purity germanium (HPGe) passive spectral gamma-ray component of the U.S. Department of Energy's (DOE) Radionuclide Logging System (RLS). The factors were derived from data collected by logging spectral gamma ray calibration standards at the DOE borehole calibration models at Hanford. Seventy high resolution gamma ray spectra were recorded during the period May 5, 1994 to May 16, 1994 for the instrument labeled RLSG2.2. The reason for the .2 extension is the fact that a new housing has been installed onto the RLSG2 instrument. This shorter housing has an effective wall thickness greater than the previous housing and therefore changes the calibration constants.

Analyses of spectra from this large data base produced an efficiency function that is used to calculate decay activities of gamma-ray emitters. This report presents these results, explains how they were derived, discusses differences between this analysis and previous calibration reduction¹⁻³, and indicates their roles in data reduction and analysis.

The calibration for specific photo peaks of K, U, and Th are not performed as in previous years. These calibration parameters are not used in the log data reduction and as such are not necessary for the log assay surveys.

Differences in calibration philosophy from previous RLS calibrations include the calibration of the tool only and not the system characterization. Casing correction based upon the wall thickness is a function of the energy of the gamma rays and the casing material and not a function of the RLS instrumentation. This year, 84 photo peaks were used for the delineation of the efficiency function, and nine of these gamma ray lines have energies below 186 keV. Another difference reported in this calibration document is the analysis for the Gross Gamma Ray (GGR) for the high purity Germanium detector. The calibration for this response is useful for normalization to previous versions of instrumentation used at the Hanford site.

It is possible to predict an observed total count rate from the estimate of the three series of emitters from K, U, and Th. This prediction is the computed GGR, CGGR. A computed GGR was also calibrated; and this is a geologic measure even in the presence of contamination. In addition, the difference between this computed GGR and the observed is a quick indicator of the presence of contaminants. The calibration models contain only K, U, and Th as do natural rock surrounding wellbores at Hanford site.

Theory Of Concentration Assay Techniques

An RLS passive gamma ray log is a depth-correlated collection of high resolution gamma ray spectra recorded as the logging tool progresses along a sequence of depths in a borehole. Each spectrum reflects the gamma ray flux in the borehole at a particular depth. A spectrum is usually depicted as a graph in which the y-coordinate of each point represents the number of detector counts at a specific gamma ray energy and the x-coordinate represents gamma ray energy.

Because nuclear decay yields gamma rays with discrete energies, a gamma ray spectrum will contain peaks that correspond to elevated counts at particular energies.

The intensity of a peak is found by calculating the count of a peak over the background, then dividing these counts by the counting time. Radionuclide concentration calculations rely on the peak intensities.

If a spectrum is recorded using known gamma ray sources, it is possible to find a relationship between the gamma ray energies and the positions of the corresponding peaks in the spectrum. This correlation is the energy calibration. After the energy calibration is determined, the gamma ray energy associated with any spectral peak can be calculated. Unknown gamma ray sources can often be identified from the energies of the emitted gamma rays. Figure 1 contains spectra collected in the SBM model which is a mixture of all three components K, U, and Th. The peaks visible in Fig.1 represent discrete gamma ray emissions. The high level of energy resolution inherent in the RLSG2.2 allow the unambiguous definition of the radioactive isotope responsible for the observed gamma ray.

Figure 1 RLSG2.2 Spectra contained in file H2MG2005.CHN

The practice of using only the photo peak information eliminates complications due to Compton scattering. The net counts in each photo peak are the counts in each peak above the continuum observed in the spectra. Analysis performed for this calibration used two commercial software applications to strip the net counts from the background continuum. First, the spectra were analyzed with an EG&G Ortec commercial

spectral analysis program named OMNIGAM^{TM(1)}. Second, low energy peaks were analyzed using commercial software by Jandel called PeakFit^{TM(2)}. When using the program PeakFit, the area is in units of Counts x keV so that the area needs to be scaled by the conversion from channels to keV.

After a specific gamma ray emitting source is identified using the value of the gamma ray energy, the decay activity, C_x , can be calculated from the observed net photo peak count rate, A_x with

$$C_x = \frac{A_x}{\epsilon(E) \cdot N_x}, \quad \text{Eq. 1}$$

where E is the gamma ray energy, N_x is the average number of gamma rays with energy E emitted per nuclear decay, and $\epsilon(E)$ is the "efficiency" as a function of energy of the RLSG2.2 instrument. Many spectroscopists would not include the formation density inside of the efficiency function as is done in Eq. 1. The self absorption of the formation would be another factor dependent upon the sample density and the efficiency function would depend only upon the instrument and its packaging. This spectroscopic approach allows the measurement of the efficiency function in the lab using commercially obtained calibrated gamma ray sources. Three examples of HPGe detector systems measuring a lab instrument efficiency are contained in the articles by K. Siemon⁴ et. al., B. Jackel⁵ et.al., and W. Lin⁶ et.al.

Spectroscopists will many times have samples of very different density and compositions. Borehole logging surveys performed at the Hanford site do not encounter a wide range of formation density values. Furthermore, the practical approach of calibrating for an instrument response in similar formations is more straightforward and less prone to operational error. Clearly, both approaches have advantages.

The instrument efficiency measured by Siemon⁴ used the same functional form as found in Jackel⁵. This functional is very complex and uses six coefficients. This form does fit RLSG2.2 data as well as the scatter in the data defines the function. TableCurve^{TM(3)} was used on the 1994 data for the RLSG2.2 in two versions. First, the data was fit as inverse efficiency versus energy, and second, as efficiency versus energy. The results of these two approaches did vary and resulted in the efficiency view being fit slightly better. Likewise, the function use by Lin⁶ was found to fit RLSG2.2 data very well. All of these functions have the character of a power law on both the high and low energy regions, with a curve over to transition between each region. Also, any of these functions fit the data within the inherent scatter of the data, (more discussion is covered in the data reduction section of this report).

The particular function used is viewed as a design decision and not an

^{TM(1)}Trademark of EG&G Ortec, Oak Ridge, Tennessee

^{TM(2)}Trademark of Jandel Scientific, San Rafael, California

^{TM(3)}Trademark of Jandel Scientific, San Rafael, California

inherent physical process for the RLSG2.2 1994 calibration. The calibration goal is to produce a methodology for measuring levels of contamination at the Hanford Site boreholes and as such an empirical function is suitable over the range encountered and calibrated. All functions studied have the same basic character from 239 to 2614 keV.

The log surveys performed in the field data reduction use the efficiency function to compute the concentrations of the gamma ray emitters. The act of calibration uses the National Institute Standards and Technology, (NIST), standard analysis of the calibration models to measure the efficiency function for the known concentrations. Equation 1 can then be recast into a different form for the calibration process. But first consider the superposition of two gamma ray emitters with the same or very nearly the same gamma ray energy. The total photo peak intensity is then written as

$$A_{total} = A_1 + A_2 = \epsilon(E) \cdot (C_1 \cdot N_1 + C_2 \cdot N_2) \quad \text{Eq. 2}$$

where subscripts 1 and 2 refer to the gamma rays from isotope 1 and isotope 2, respectively. For example, K gamma ray energy is 1461 keV, and Th series results in a gamma ray with 1459 keV. Although the absolute gamma ray activity of the Ac^{228} in the Th series is different from the absolute activity of the K^{40} gamma ray, the result is an interference that is indistinguishable using only the gamma ray spectra at this energy. Adjusting Eq. 2 to

$$\epsilon(E) = \frac{A_x}{\sum C_i N_i} \quad \text{Eq. 3}$$

where A_x are all the counts at the average energy x and each individual emitter concentration and associated absolute intensity are C_i and N_i , respectively. Note that if some isotopes are ignored then the sum in the denominator of Eq. 3 will be too low and the resultant estimate of ϵ will be too large. Thus an indication of a high ϵ data point from the observed trend is that there are some interferences not accounted for from this peak.

RLSG2.2 Calibration Data Reduction

The DOE spectral gamma ray calibration models located at Hanford were measured with the RLSG2.2 instrument during May 6 to May 16, 1994. This data has been archived and the analysis for calibration performed according to the procedures stated in EII 11.1, Appendix B, Revision 2, of WHC Environmental Investigations and Site Characterization Manual (WHC-CM-7-7).

The individual spectra were analyzed using the commercial program OMNIGAM^{TM1} for all the photo peaks 186 keV and higher. The low energy gamma ray intensities were obtained using PeakFit^{TM2}. Under normal data collection setup, the low energy peaks cannot be analyzed since the peaks are closely spaced and have insufficient channel description. Therefore, separate spectra were collected in the SBU and SBT models with an electronic gain

4 times the normal setting. Thus the energy ranged up to near 650 keV for 4096 channels in these special cases instead of the usual 3000 keV. Figure 2 contains data from the SBU model under these two gain settings. As can be seen, the low energy peaks do not have enough character or data density to properly strip the net count in the normal data collection mode. The special data collected in the same models with the expanded gain setting allows the use of commercial peak fitting software to obtain the net gamma ray intensity. Note that the peak at 77 keV has only two channels in the logging mode data collection scheme and there are 11 data points in the data collected especially for this calibration study.

Figure 2 Comparison of data density for the normal gain setting and the special 4 times value.

The log survey does not collect data in this expanded gain mode. Therefore, this technique cannot be used for a survey. However, the use of this data during calibration allows the determination that the 186 keV point is in error, and can be eliminated from the fitting procedure. Furthermore, the application of a better behaved function can also be used instead of the apparent best fit. This fact is significant since the efficiency data is so poorly behaved, (this point is expanded later). The characterization of the efficiency data as poorly behaved is relative. The accuracy for the RLS at 10 1000 sec spectra in the calibration models is much better than the other sources of error in the calibration process.

The possibility of some systematic error between stripping techniques of both OMNIGAM^{TM1} and PeakFit^{TM2} was investigated, but the results verify no difference. The U series gamma ray at 609 keV is prominent in both spectra collection schemes. Thus, the two software packages were used on this gamma ray in the expanded gain spectra with the results of 2/10 of 1% difference between measured count rates.

Figure 3 SBT model calibration spectral fitting results for expanded gain data.

Therefore, use of the low energy gamma rays fitted by PeakFit^{TM2} can be used with the differently stripped high energy peaks.

Figure 3 displays some of the data taken in the SBT model at expanded gain. The results of the spectral fitting of Gaussian peaks is shown, (individual peaks are shown at the bottom of fig. 3, and the solid line is the sum of peaks and background). The five peaks used to fit this section of data from 85.5 to 96 keV could have been expanded to include a sixth peak centered about 95 keV, the shoulder of the highest peak in fig 3. However, the smaller peaks are subject to large statistical errors and are only used to obtain a good fit for the more intense peaks. Only the most statistically accurate peaks were used in the efficiency measurement as discussed in the previous section. The resultant total number of usable low energy gamma rays obtained with this technique is nine for both the SBT and SBU models. (Note that the individual peaks shown in Fig. 3 are plotted on a separate and relative scale without Compton background.) Clearly, the overlapping of the many low energy gamma rays require a simultaneous fit of several Gaussian peaks in order to arrive at an accurate gamma ray intensity for any one peak.

The SBU, SBT, SBK, and SBM models have been designated by DOE as calibration standards for passive gamma ray logging. Each model contains a test zone that consists of concrete with a uniform distribution of K, U, and Th. The decay activities or concentrations of the test zones are listed in Table I. Other model parameters appear in Steele and George⁷.

Table I Gamma Ray Calibration Models

Zone	K (pCi/g)	U (pCi/g)	Th (pCi/g)
SBU	10.72	190.52	.66
SBT	10.63	10.02	58.11
SBK	53.50	1.16	.11
SBM	41.78	125.79	39.12

Calibration gamma ray spectra were recorded with the tool suspended and held stationary in the model test hole at a depth such that the center of the HPGe detector was at the middle of the test zone. Calibration data were taken under the following borehole conditions, which are defined as standard:

borehole diameter 4.5 inches
 borehole fluid air
 casing none.

The calibration models located at Hanford were originally installed by DOE as secondary standards at the Spokane site during the 1970's. These models were then moved to the Hanford Site in August 1989. On October 3, 1992 Kaiser Engineers Hanford began the installation of the models in their permanent positions. During this shipment date and installation date, the models were stored dry. Possibility of changes in Radon containment initiated a study by Koizumi⁸ to measure the status of the concentrations. The conclusion of this report is that the models had not changed since the upgrade in concentration assignment by Steele and George⁷. All previous RLS calibration procedures used the models located at the Grand Junction facility. This year is the first year to use only the Hanford models for RLS calibration.

The OMNIGAM^{TM1} program automatically stripped photo peaks from all the

spectra for a given list of target gamma rays. All net count rates were then dead time corrected by using the Ortec electronic measure of Multi Channel Analyzer, MCA, elapsed time. The values for C_i , (EQ. 3) for each of the gamma rays are listed in Table I. The respective values of N_i , (EQ. 3) were obtained from Erdtmann and Soyka⁹. Using Eq. 3, the computed efficiency values for some of the observed gamma rays are shown in Fig. 4. The preselection for these data was the criteria of relative statistical error of 10% or better for an individual spectra. For example, several U and Th series gamma ray peaks were stripped from the K model spectra, but the relative error for these gamma ray intensities were 20 to 50%. Inspection of these individual peaks in the spectra revealed no peaks noticeable above noise of the background continuum. The stripping function was blindly getting peaks from the noise.

Figure 4 Efficiency data for all four model zones at Hanford calibration facility.

There are several special cases that must be discussed. The gamma rays at 186, 727, 911, and 1001 keV are clearly off the expected trend for a typical spectroscopic efficiency function. The expected character of an efficiency function is a power law function rising with energy at the low end and falling at the high energy region. Plotting on a log log scale then shows two straight lines with a curve over in the connecting region. Figure 5 is a log log plot of 1994 RLSG2.2 data, power law fit (excluding 186, 727, 911, and 1001 keV points), and the dotted curve is the quoted efficiency function by Lin and Harbottle⁶ normalized to the two 186 keV data points. It is clear that the 186Kev data points do not follow a power law character with the majority of the high energy data. Likewise, the 727 and 911 keV data do not match the major trend. Inspection of the spectra for these data did not reveal any difficulties with peak stripping or double interferences. The 186 keV data has the gamma ray interference

Figure 5 Log log plot of data, power law fit (solid line), and Lin and Harbottle⁶ referenced efficiency function (dotted line).

from both U^{238} (186.0 keV) and U^{235} (185.7) computed as in Eq. 3. In fact, the Th^{232} (184.6Kev) was also included with no major change. As pointed out earlier, any efficiency data plotting too high may have physical sources present but not added into Eq. 3.

The method used (Eq. 3) for accounting for gamma ray interferences yields accurate results for many other gamma rays. There were 24 peaks used that had contributions from two or more isotopes. Only the 186, 727, 911 keV peaks show a deviation from the power law trend of the rest of the data. These three peaks do have contributions from two isotopes.

A second escape mechanism from a higher energy gamma ray cannot contribute to the 186 keV peak. The threshold for escape mechanism is the sum of two electron masses or 1022keV. However, the 727, 911, and 1001 keV peaks could potentially have contributions from a gamma ray 511 keV above each peak.

The 727 keV peak was individually inspected for all the spectra with no noticeable reason for a discrepancy. It is a nice single peak well above background. There is a peak at 1238 keV from Bi^{214} that theoretically could produce a second escape into the 727 keV peak. However, there are two reasons this phenomena cannot explain the mis-match of this peak from the trend of the other gamma ray points. First, the probability for second escape at this energy is extremely low. Second, the efficiency plots too low from the trend and thus would plot lower still if more counts were in the peak from that already assigned.

The efficiency data from the 911 keV peak has strange behavior. There are two data points that have a significant peak at the 911 keV, the SBT and SBM models. Thorium is the dominant contribution to the 911 keV by the daughter isotope of Ac^{228} . A uranium daughter (Tl^{210}) has a gamma ray at 910 with an absolute intensity of 3.00 per 100 decays listed in Erdtmann and Soyka⁹. The strange behavior is that the SBT model results are consistent with the trend of the other data, while the SBM results plots significantly low. If you assume no contribution from the Tl^{210} gamma ray, then both model results yield an efficiency of .0093, only 11% low from the fit results. The photo peak width would not fully resolve gamma rays this close (910 and 911) at these relative intensities. Therefore, both must be included in the efficiency calculation. The error is not quoted in Erdtmann and Soyka⁹ for the absolute intensities listed. The possibility of errors in the assigned absolute intensities cannot be ruled out. More discussion of this aspect is covered later, when the systematic efficiency deviation is covered.

The efficiency data from the 1001Kev photo peak was not used in the least squares fit shown in fig. 5. The major subject of the paper by Siemon³ et. al. is that the Pa^{234m} gamma ray at 1001 keV has some disagreement concerning the absolute intensity. In an internal memo from Koizumi¹⁰, the RLS is recommended to adopt the new value of .83 gamma rays per 100 decays, since the RLS calibration data from Grand Junction models supports the quoted value. This new value (.83) was used in the analysis, however, there remains an average of 16% deviation from the fit results.

The conclusion of these special cases is that they were not included in the fit for the efficiency function. There is no concrete reason that could be arrived at in order to eliminate these gamma ray peaks from the

definition for the efficiency function. The only rational is the character of a power law function that is demonstrated to exist for referenced HPGe systems. This functional character can be coupled with the poor behavior of the data even with the elimination of the significantly deviated points to suggest that the quoted values of absolute intensity are questionable. The results are purely empirical. The resultant function is

$$\epsilon(E) = \frac{1}{a \cdot E^{\alpha} + b \cdot E^{\beta}}, \quad \text{Eq. 4}$$

where a , α , b , and β are constants adjusted by the non linear least squares fit.

The log survey data collection system does not allow the proper stripping of data points with an energy below 110 keV. Therefore, the use of Eq. 4 is not totally applicable. That is, the contribution from a and α is negligible for all energies above 186 keV. Operationally, it is possible to only use the single term in the denominator and therefore only two coefficients to determine contaminant concentrations. (Note that any singlet peaks between 186 and 110 keV could be analyzed with Eq. 4. The possibility now exists to extend the calibration to include Pu^{239} , Pu^{240} , U^{235} , Am^{241} .)

The resultant values for these constants are:

$$\begin{aligned} a &= 10.516 \pm 1.74 \\ \alpha &= 0.32325 \pm 0.0232 \\ b &= 2.36 \cdot 10^{10} \pm 2.8 \cdot 10^{10} \\ \beta &= -4.217 \pm .277 \end{aligned}$$

This function (Eq. 4) is the solid line in fig. 5. Thus the efficiency can be computed using energy in keV and the above constants. Then use Eq. 1 with A_x in counts per second and N_x in gamma rays per 100 decays to arrive at the concentration of the unknown, C_x (in pCi/g) from any spectra obtained by the RLSG2.2 instrument in a borehole that is air filled (no casing). Any environmental corrections¹ such as casing thickness, fluid filled borehole, or borehole diameter must be applied to the count rate, A_x , before use of Eq. 4 and Eq. 1 to arrive at the formation concentration of the gamma ray emitter. Likewise, a dead time correction must be performed first, and is usually done with the electronic elapsed time.

There are two additional models at the Hanford site, the SBA and the SBL. These were initially designed for calibrating gross gamma ray logging. They are constructed like the U model, SBU. Later in this report, they will be used for just this gross gamma ray calibration. As a verification of the calibration of the efficiency function, data in the SBA, SBL, and SBU were put through the resultant efficiency function in order to ascertain the concentration of U. In the gross gamma mode only, the SBU is assigned a U concentration of 185 pCi/g, this is different from the 190.5 pCi/g used by this same model in the spectral assay mode⁷. The reason for this difference is unknown. However, it is assumed that the assignment of the U concentrations for the gross gamma (SBA and SBL) would also have this 2.7% difference for the spectral mode of analysis such as is being performed with the RLSG2.2 data. Therefore, this report used 1.027 times larger assigned concentrations for the SBA and SBL than quoted

by Steele and George⁷.

The average concentration from 17 gamma rays for each of the U models is given in Table II. Also, the assigned⁷ values are given in Table II. The observed scatter quoted in column 2 of Table II is the root mean square of the measured concentrations from the 17 gamma peaks used. This error has some interesting characteristics, which will be discussed in the next section.

Table II Results of RLSG2.2

Zone	RLSG2.2 (pCi/g) Ra ²²⁶	Spectral Grand Junction (pCi/g) Ra ²²⁶	Grand Junction (pCi/g) Ra ²²⁶
SBU	192.7 ± 5.3	190.5	185
SBA	64.0 ± 1.9	62.8	61.2
SBL	341.1 ± 13.1	332.7	324

Error Discussion

The total error for the calibration coefficients can be estimated from the individual contributions as well as observed in the scatter of the 84 data points used. Consider Eq. 3 (copied here for reference);

$$e(E) = \frac{A_x}{\sum C_i N_i} \quad \text{Eq. 3}$$

There are four error processes on the right hand side of Eq.3 listed above. First, the error inherent in the summation of the denominator can be considered the proper selection of all the contributors to each photo peak intensity. All daughter isotopes with an absolute gamma ray intensity of 0.1 per 100 decays or greater were used. The second component of error is the known value of each concentration, i.e. C_i . The errors quoted for the concentrations⁷ of U, Th, and K are about 1.5% one standard deviation. This is based upon NIST standards. Third source of error is the values used for the absolute intensities, N_i . No estimate of these errors are available. Fourth source of error is the measurement of the photo peak intensity A_x from the spectra. The individual peaks used varied from 3% relative error to 0.52% for the smallest, (using 10 repeat spectra from each model). The error bars shown in fig. 4 are this last category of error for the calibration analysis contribution from the error for photo peak intensity.

In general, the observed scatter of the data for Eq. 3 is much larger than the error in the estimate of the photo peak intensity, (refer to fig. 5). The conclusion is that one of the other three components of error source is significant. As discussed briefly before, the possibility of second escape peaks falling into a lower photo peak and sum peaks from various lower peaks falling into a higher photo peak could contribute to error. However, for the energy range used for the calibration (2614 keV and less), the probability for these physical mechanisms is small and

insignificant. Likewise, interference gamma rays of different peaks than these problem peaks (with nearly the same energy), have been correctly accounted with the summation in the denominator of Eq. 3. Photo peak stripping techniques were considered by using two different commercial programs, with excellent agreement. The most significant error in photo peak intensities is the statistical errors in stripping from the continuum background.

The observed scatter in the concentrations computed for the RLSG2.2 data in the three U models is four times larger than what could be called a reproducible error. The deviations observed for each of the 17 gamma rays used is reproduced in each of the three models. For example, the concentration resulting for the 609 keV peak is low from their average by 4.2%, 3.0%, and 3.7% in the SBU, SBA, and SBL, respectively. The repeatability in the different models has a precision that is much better for a given gamma ray than for the collection of gamma rays. Consider the 767 keV gamma ray where the difference is a consistent 3.6% high for the three models. Figure 6 shows a plot of the relative difference from each mean for each model. The relative difference for each model is defined as the percentage difference from the mean concentration for one U model. The deviation of all individual points shown in fig. 6 is 3.2% and the average deviation for each set of three Uranium measurements for all gamma rays is 0.8%, a factor of four times smaller.

One possibility for the points reproducing better than the observed scatter in all the data points is a systematic error for each gamma ray in the absolute intensity used from Erdtmann and Soyka⁹. Also this error would need to be different for each of the observed gamma rays, thus accounting for the observed scatter from energy to energy. The Ortec software included reference libraries and absolute intensities based upon the Evaluated Nuclear Structure Data File (ENSDF)¹¹ from Oak Ridge. This library lists the U235 gamma at 185.7 keV with 57.5 gamma per 100 decays. Using this value causes the 186 keV efficiency to plot on the power fit with no noticeable deviation. However, several other gamma rays, (in particular the 2614 keV) show significant deviations from the general

power law trend. Therefore, using either library results in the exclusion of selected points from the general trend of the vast majority of the data following a power law character. Likewise, the observed 3.2% scatter exists using either library. In addition, the absolute intensities quoted in the two libraries do differ in values.

The calibration error quoted in this report for the coefficients in the efficiency function were obtained by the commercial software PeakFit^{TM2}. The error obtained from this type of statistical non linear least squares fitting can be mis-leading. Frequently, the standard error is more a representation of how well the data fits the function and not the inherent error in the coefficients. For example the reproducibility of any given point in this data set is much better than the error computed using the propagation of error from the coefficients used. Also, it must be remembered that the discussion here is the error in the calibration process and not any given log survey.

An alternate method of perception can be envisioned. The multitude of efficiency data points is not well behaved in that they scatter more than the statistical error. Furthermore, the large number of points forces a least squares fit to a function with much better precision than the observed scatter in the data from this resultant fit. However, no analysis technique can then estimate this reproducible level of error. The worst case error is quoted in the coefficients listed on page 9 for the efficiency fitting algorithm. Thus the error listed with these coefficients is representative of the scatter in the data from the fit results, and is much larger than the reproducibility of the RLSG2.2 system.

One alternative is to produce an estimate of the overall reproducibility of the RLS system in general. Since 1991 there have been many RLS calibrations both in Grand Junction and Hanford, and this includes several different tool configurations, (crystal size and housing thickness). After each calibration of each instrument, the readings in the U model could be assayed for the average of all 17 prominent gamma ray lines. This value for each calibration set can then be used to observe the repeatability of the RLS system to yield the same answer year to year and tool to tool. This estimate would better represent the reproducibility of the system but not the absolute error of the calibration.

The absolute error of calibration is mostly a function of the accuracy of the models from the NIST standards, and the scatter of the data. These combine to yield an estimate of about 3.5% for the usable high energy range of 200 to 2614 keV. This estimate is determined from the 3% observed scatter from the verification measurements shown in fig. 6 and the quoted error of 1.5% in the assigned values⁷ of the models.

Gross Gamma Ray Calibration

Geologic Gamma Ray

The natural gamma ray log has been important to earth scientists since the first log in 1938 for identifying geologic changes in various strata penetrated by the borehole. Correlating geologic conditions with plume distribution is just as important today at Hanford because of the geologic strata influence on migration. Without geologic information, driving mechanisms, directions of movement, speed of movement, and species dependent mobility, predictions of movement cannot be accurately defined.

The site specific application of the gross gamma ray is both a geologic use as well as a quick (screening) estimate for contamination. The calibration of the gross gamma ray can then use any standard such as API, Equivalent Parts Per Million Uranium, or a historical count rate. The issue is a design decision and not restricted to a particular physical process. The chosen standard is pCi/g of Ra^{226} , since these are the units for the calibration models. The historical continuity is deemed the most valuable aspect, but the geologic use is not precluded with this choice.

The geologic use of natural gamma ray logs at the Hanford Site is complicated by the presence of man made radionuclides. A method of computing the natural gamma activity (natural rock gamma rays) from the borehole signal containing both man made and natural gamma rays has been developed. This type of log product uses any type of spectral data and should be ready for implementation in two months. It is noteworthy that this product does not require new instrumentation, but simply reprocessing of the data.

The use of this predicted natural gross gamma ray for the screening instrumentation has an additional benefit. The comparison of the predicted natural gross gamma ray and the observed gross gamma ray will flag the presence of total levels of man made emitters.

Back Calibration

The WHC Geophysics Services has recently demonstrated the ability to back calibrate older log data using the RLS information. This capability to normalize older instrument responses adds much more history to a vadose monitoring program. This significant technical advance is valuable in leveraging the time analysis of the slow natural movement of contaminants in the vadose zone.

In theory, a comparison of older log data with data from new instrumentation is possible by normalization of the calibration results. However, the older data does not necessarily have the proper calibration procedure for this type of normalization. In addition to the possible problems with the calibration of these older instruments, the potential for instrument drift or change after calibration presents problems for normalization. Clearly, the determination of contaminant movement or non movement is not possible if two different instrument produced data cannot be matched in their responses. The action of different decay rates for common contaminants creates another variable in the definition of contaminant movement.

It is possible to use the log data to determine a normalization factor under some circumstances. The first variable to be defined is the different zones of different decay rates. This can only be performed with RLS data that can identify the species components. Next, zones of some dynamic range must exist in order to match log response from the log data. Not all wells and their log data will be subject to this log analysis type of back calibration. However, the extension of time lapse data to issues of contaminant movement is beneficial to a vadose monitoring program.

Figure 7 Normalization of 3 different logging instrument responses for gross gamma ray.

Figure 7 contains an example of a back calibration for three logs. The three instruments used were a) RLS HPGE, b) RLS version of NaI, and c) a gross count NaI used by PNL. Also, multiple well log analysis using older data, that has been validated with the RLS, can assist in the delineation of past incidents from current conditions.

Gross Gamma Ray Analysis

The same spectral data taken to calibrate the RLSG2.2 for the concentration assay is used to provide the gross gamma ray activity. The only usable models are the Uranium only models of SBA, SBU, and SBL. These models and their assigned concentration⁷ are listed in Table II. The calibration of the computed gross gamma from the KUT concentrations used all six models. The gross gamma count rates for all models are listed in Table III.

Table III Gross Gamma Ray Calibration

Model	Gross Gamma Ray Observed (Cts/sec)	Assigned Ra ²²⁶ (pCi/g)
SBA	3379.3	61.2
SBL	18477	324
SBU	10207.4	185
SBT	3985.7	Not Applicable
SBK	253	Not Applicable
SBM	8991.1	Not Applicable

The gross gamma ray count rate is the total count rate for all energies. The computed gross gamma calibration can only use the three models SBA, SBL, and SBU, since the

other models have significant amounts of other sources besides Uranium. However, the computed gross gamma ray uses the observed total count rate and all the models. Therefore, the Table III contains all observed total count rates but only the assigned values for the first three models.

The resultant linear expression for the GGR response is shown in Eq. 5

$$C = C_0 + m \cdot GGR, \quad \text{Eq. 5}$$

where C is the concentration of Ra^{226} (pCi/g), GGR is the RLSG2.2 total count rate (cts/sec, dead time corrected). The coefficients obtained are:

$$C_0 = 4.95 \pm 5.38$$

$$m = 0.0173 \pm 0.0004.$$

The conversion to other units such as equivalent parts per million U (EQppm U) are discussed in Koizumi¹, et. al. Once this gross gamma ray calibration procedure is established, the application to back calibration is much easier for future changes in instrument response. The data and resultant least squares fit are shown in fig. 8.

The computed gross gamma ray, CGGR, uses the superposition principle. Each source (or identifiable series) produces a characteristic spectra and thus a characteristic value for the total observed counts. Simultaneous solution for a given total observed total from the three sources, (KUT) for each of the six models was performed. The relation used is shown in Eq. 6,

$$CGGR = ak \cdot CK \cdot (1 - 0.0292 \cdot \frac{CT}{CK}) + au \cdot CU + at \cdot CT, \quad \text{Eq. 6}$$

where CGGR is the computed gross gamma ray count rate, ak, au, and at are the fit coefficients, and CK, CU, and CT are the count rate of the net photo peaks for 1461, 609, and 2614 keV respectively. The interference from the Th and K lines is accounted for by the 0.0292 of the observed Th count rate. If there are no other sources in the formation, then, the CGGR should be equal to the observed total count rate. Any difference between these two quantities is a flag for man made gamma ray emitters. The results are:

$$ak = 35.1456$$

$$au = 100.009$$

$$at = 221.344.$$

The observed standard deviation of the computed gross gamma ray using these coefficients from the observed in the six models was 1.7%. Thus the accuracy of applying Eq. 5 is near 2% compounded with the error for the

three peak intensities. Generally, the statistical error for the observed gross gamma ray will be much less than the error for the computed gross gamma ray. Therefore, the error in the computed gross gamma ray difference from the observed gross gamma ray is all due to the computed gross gamma ray. This is important for setting a threshold limit for the man made indicator. The recommended threshold is 5% difference for any given spectra, but a log response with a consistent difference, (several points consecutively), above 2% is significant.

A casing correction is presently not possible for this computed GGR, since the Compton downscatter is a significant component of the observed total count rate. However, a log analysis computation can be applied to cross correlate the two total count rates in clean zones for a particular casing.

The calibration parameters found are sufficient for borehole surveying. In fact, the low energy region has been extended over previous measurements. The highly empirical nature of this calibration is quite sufficient for the goal of assaying the levels of contaminants in the formations encountered at the Hanford site.

Future Recommendations

The following list describes recommendations for action concerning the calibration and standardization of the RLS for contaminant surveying.

The unanswered physical questions concerning the crucial 186Kev photo peak leads to recommendations of making further studies. A laboratory measurement of the efficiency function for only the RLSG2.2 instrument be performed. This measurement would improve the support of the calibration of the RLS in general.

The calibration models should be scanned azimuthally to check for homogeneity. Likewise, centered versus eccentered should be measured to document the character of the models.

The observed systematic deviation of the measured efficiency function must be investigated and corrected. One possible support for the explanation would be the measurement of a tool only efficiency function with sources that do not have so many interferences. However, the Thorium spectra also displayed a similar deviation dependent upon the particular gamma ray viewed. Likewise, other literature sources must be investigated for the absolute intensity of these gamma rays for Thorium and Uranium.

Future calibration must extend the efficiency function to higher energies, if the tool is ever combined with a neutron generator. The neutron induced spectroscopy require efficiency values at higher energies than 2.6MeV. The lab measurement (tool only) could help this issue. Likewise, the low energy calibration can be further refined.

Generate a procedure for calibration. The goal is to perform a calibration and update the logging tool data base with the appropriate coefficients and documentation without the generation of a separate calibration report each time a calibration is performed. A simple form with appropriate signatures, coefficients, and dates are all that is

required. Joint effort between Grand Junction and Hanford technologies would produce a better standardization of the calibration procedure.

Generate a computed gross gamma ray for each survey and compare to the observed. This requires a casing correction to be applied to each channel of the spectra. Therefore, a different casing correction algorithm is required.

The photo peak casing correction should be separated from the calibration since it is not a function of the tool. The casing correction transforms the gamma flux from one side of the casing to the other side of the casing. It should be possible to measure the casing correction in the lab with overnight runs, saving day time for borehole surveys. Likewise, extend the energy range of the casing correction to both lower and higher energies.

Develop a Strontium detector and establish threshold limits for the observed bremsstrahlung from the beta activity. Conaway¹² has described a method to produce composite spectra for each emitter. That is the entire gamma ray spectra is computed from the intensity of each single photo peak. The total of all these contributions can then be subtracted from the observed to show the Strontium continuum spectra if it is present. A straight forward computation would be to take the same approach as discussed for the computed gross gamma ray. Simply compute a gross for each identified radioactive isotope and subtract from the total observed.

Calibration of the strontium detection requires several items. First, the casing correction must be applied to the entire spectrum. Second, the threshold limits must also be calibrated. Third, photo electric absorption must be corrected for log survey data, especially for the casing correction algorithm.

The dead time correction used appears to fail at the higher count rates. The validation of the correction must be performed. The limits for the correction to be accurate must also be measured.

An algorithm to use multiple peaks when possible should be field tested. Initial tests did not result in any observed enhancement. Thus, the reason for this result should be studied and corrected.

It is possible to determine if the gamma rays are sourced throughout the formation or concentrated just outside of the casing. The simplest test is to use multiple peaks from the same isotope to measure their relative intensity. A formation distributed source will have more intensity loss for the lower energy peaks. Clearly, the accuracy will be driven by the casing correction. In addition, isotopes with only one gamma ray such as Cs¹³⁷ must be analyzed by other means. It is possible to measure the photo peak intensity relative to the Compton edge for the same kind of information. This data analysis procedure needs to be calibrated.

Deconvolution for each of the passive gamma ray logging systems needs to be developed and implemented for monitoring surveys at the Hanford Site. Thin zones for one tool may not be a thin zone for another tool thus, the two tools would read differently and possibly be mistaken for time lapse movement.

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