Spectrum Shape-Analysis Techniques Applied to the Hanford Tank Farms Spectral Gamma Logs

May 1997

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**Spectrum Shape-Analysis Techniques Applied
to the Hanford Tank Farms Spectral Gamma Logs**

May 1997

MASTER

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Executive Summary

Gamma-ray spectra acquired with high-energy resolution by the spectral gamma logging systems (SGLSs) at the U.S. Department of Energy Hanford Tank Farms, Richland, Washington, are being analyzed for spectral shape characteristics. These spectral shapes, together with a conventional peak-area analysis, enable an analyst not only to identify the gamma-emitting species but also to determine in many instances its spatial distribution around a borehole and to identify the presence of the bremsstrahlung-producing contaminant ^{90}Sr . The analysis relies primarily on the results of computer simulations of gamma spectra from the predominant radionuclide ^{137}Cs for various spatial distributions. This log analysis methodology has evolved through an examination of spectral features from spectral logs taken at the SX, BY, and U Tank Farms at the Hanford Site. Initial results determined with this technique show it is possible, in most cases, to distinguish between concentrations of ^{137}Cs localized to the borehole region and concentrations of ^{137}Cs either uniformly distributed or remotely located in the surrounding vadose zone. When the ^{90}Sr activity is sufficiently large, this method can also be used to identify the presence of ^{90}Sr , with or without the presence of ^{137}Cs . Work is continuing by experimentally measuring shape factors, incorporating spectrum shape processing in routine log analysis, and extending the techniques to additional radionuclides.

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1.0 Overview

The Tank Farms Vadose Zone Characterization Project at the U.S. Department of Energy (DOE) Hanford Site was conceived as a borehole logging effort using the existing array of monitoring boreholes within several tank farms to collect spectral gamma logs that identify radionuclides and their activities as a function of depth. These data are then used to develop horizontal and vertical distributions of the gamma-emitting contaminants in the unsaturated sediment surrounding the underground single-shell waste tanks.

Gamma peak intensities measured by spectral gamma logging systems (SGLSs) are converted to contaminant activity by the use of calibration constants determined from measurements in uniform distributions of the naturally occurring gamma emitters potassium, uranium, and thorium (KUT) (DOE 1995a). This uniformity must exist both radially away from the borehole and axially with depth. Because gamma rays do not transport far in earth materials, the logging tool response corresponds to the response for a uniform source distribution if the radial extent is about 30 centimeters (cm) (11.8 inches [in.]) or greater and the axial extent of the contamination is about 40 cm (15.7 in.) or greater. These distances are related to gamma energy and to the fraction of the uniform response that is required. Spectral gamma logs show when the axial distribution lacks uniformity, but radial nonuniformity cannot be detected from only gamma intensity. If the contaminant distribution encountered while logging is not radially uniform, the predetermined calibration constants will not be appropriate and computed activities will be incorrect. A technique is needed to identify radially nonuniform contamination. In addition to "flagging" depth intervals that correspond to incorrect activity concentrations, this identification also would help an analyst understand the nature of the measured contamination and perform the difficult transition from depth distributions of contaminants to interpolated horizontal distributions.

Gamma rays from a monoenergetic source that traverse a medium in which scattering and absorption occur produce a scattered continuum of energies below the source energy. The shape of this continuum and its magnitude relative to the unscattered source radiation are sensitive to the intervening material and to the distribution of the source. Other researchers observed these spectral shape effects and exploited them in developing tracer logs for the petroleum industry (Gadeken et al. 1991; Gartner et al. 1990). Enhancement of the low-energy component of the gamma spectrum from ^{60}Co spectral gamma logs was recently interpreted as indicating the contaminant was inside the casing for a Hanford monitoring well at a crib disposal site (Gadeken et al. 1995). Early work by Pacific Northwest National Laboratories (Nielson et al. 1977) suggested that the ratio of scattered continuum counts to the counts in the ^{137}Cs photopeak could be used to determine if the contaminant was located near the borehole or distributed throughout the sediment surrounding the borehole. On the basis of this earlier work and the successful use of spectral shape measurements in tracer logging, computer simulations were suggested to determine if this effect can be exploited with SGLSs to determine ^{137}Cs source distribution and to identify the presence of ^{90}Sr .

This study of the development of gamma spectral shape-analysis methods focuses on the contaminants ^{137}Cs and ^{90}Sr . Future enhancements will include other contaminants, such as ^{60}Co

and ^{154}Eu . Three distinct ^{137}Cs source distributions were considered in this study: (1) contaminant confined to the inner or outer surface of the borehole casing, (2) contaminant uniformly distributed in the sediment, and (3) contaminant located remotely from the borehole in the sediment. Because experiments that measure the required spectra are difficult to perform, reliance was placed on the use of computer simulation models. Some of the simulation results were compared to experiments with known sources and distributions, but there is still a critical need for benchmark comparisons to validate the computer simulations. ^{90}Sr bremsstrahlung spectra were simulated, but the existing models are crude and the results are limited in usefulness. However, these results proved useful in estimating the detection limits for ^{90}Sr and in providing guidance in developing spectral shape-analysis techniques that indicate both the presence of ^{90}Sr and the nature of the ^{137}Cs source distribution.

This report presents descriptions of the computer models used in simulating SGLS spectra for a variety of borehole and source distribution conditions and the modeling results. To interpret locations of contaminant distributions, a spectral shape-analysis methodology was applied to logging data from boreholes at the Hanford SX Tank Farm.

2.0 Computer Simulation of Gamma-Ray Spectra

2.1 Code and Model Description

Simulation models for the response of an SGLS to gamma rays and bremsstrahlung produced by ^{90}Sr were developed with the MCNP (Monte Carlo neutral particle) computer transport code. Los Alamos National Laboratory scientists wrote this code. It has been extensively benchmarked (verified by comparison with experimental results) and has been used throughout the world for many years to simulate a wide range of radiation transport problems. Version 4A of MCNP, as documented in Briesmeister (1993), was used in this study. A relatively new feature of MCNP introduced in version 4, the pulse-height tally, was used for the SGLS simulation models. Because this tally has not been benchmarked as extensively as the remainder of the code, benchmark experiments are needed to support and verify the SGLS models. A limitation of this tally in MCNP is that most variance reduction features available with the code cannot be used. Only source biasing can be employed without causing the tally to be incorrect. For this reason, the present MCNP models of the SGLS tool produce poor statistical precision in simulated spectra for gamma sources that must be transported long distances, in terms of mean-free paths, through a medium to the detector. Therefore, the models produce statistically poor spectra simulations for gamma sources at radial distances greater than about 50 cm (19.7 in.) from the borehole. Other simulation models that employ the discrete-ordinates method are being developed for these so-called "deep-penetration" problems.

A reasonably detailed geometric model of an SGLS probe was developed for simulation models in this study. Figures 1, 2, and 3 present three views of the model geometry used in the MCNP simulations. The 3.38-inch (in.)-diameter logging tool is fully eccentric against the wall of a cylindrical iron casing that has a 6.25-in. outside diameter and a 0.313-in. thickness. The borehole fluid is air, which is the case for virtually all SGLS logs collected to date. The formation surrounding the borehole is dry silica with a bulk density of 1.8 grams per cubic centimeter (g/cm^3). Other formation compositions were considered, but silica is reasonably representative of most earth formations and was the nominal formation material used in this study. Simplifications to the actual tool design achieved a reasonably simple simulation geometry. Representation of the material around the high-purity germanium (HPGe) detector is particularly important to produce an accurate model of SGLS spectral gamma response. Response sensitivity to tool materials depends on the gamma source distribution: point sources near the detector will produce the greatest sensitivity, and sources distributed in the formation will produce the least sensitivity. For this simulation, the thinned section of the tool housing surrounding the detector has a thickness of 0.05 in. The bull nose shape chosen for the model is an approximation but contains about the correct amount of stainless steel. The sections above the detector also are approximations, composed of homogenized mixes of the major materials with an assumed dilute bulk density.

Figure 1. Vertical Section of MCNP Simulation Model Geometry, Including Tool and Borehole Axes

Figure 2. Magnified Vertical Section of MCNP Model Geometry Showing Tool Details

Figure 3. Magnified Horizontal Section of MCNP Model Geometry

Spectra were measured with point sources at several positions along the surface of the tool and were compared to simulations for differing material densities above and below the detector to determine the sensitivity of spectral shape to those parameters. The variations of these material bulk densities required for good agreement with the measured point-source spectra did not affect the simulated spectra for formation-distributed sources by more than a few percent.

Figure 1 is a schematic that is truncated radially and axially to show the important "near-field" features of the simulation model. The formation (silica) actually extends 75 cm (29.5 in.) radially and +134 cm (52.8 in.) and -100 cm (39.4 in.) axially. Surrounding the model is a void region where photons are "killed" if they reach this area.

Figure 2 shows the near-detector region. The thinned section of the stainless steel housing is 18-cm in length; the detector position within the housing was determined by the detector centerline shown on tool housing drawings provided by the manufacturer. Ortec, Inc. (the supplier of the detector, cryostat, and electronics) furnished the detector dimensions, dead layer interval, and inner core size. The cold finger and other materials connecting the detector to the nitrogen dewar are approximated as a dilute mixture of Fe and Cu. The borehole area around the tool is filled with air at atmospheric pressure. The region identified as Fe on Figure 1 is the borehole casing.

Figure 3 shows a horizontal section of the model geometry, confined radially to the near-detector region. This section transects the midplane of the HPGe detector at an axial position of 0 cm. Two thin (1-millimeter [mm]-thick) annular voids are positioned at the inner- and outer-casing surfaces to simulate gamma sources on either the inside or the outside surface of the casing wall. The circles imbedded in the formation, concentric with the borehole, divide the formation into annular sections for use by certain routines in the Monte Carlo software.

Simulations were conducted with many different source geometries, all with cylindrical symmetry around the borehole axis. Concentrations of ^{137}Cs were distributed within a 1-mm annulus at the inner casing wall, at the outer casing wall, uniformly throughout the formation region, or in 1-cm-thick formation annuli at various radial distances from the borehole wall. Additional source distributions included ^{137}Cs from the borehole wall to various radial depths into the sediment. Simulations of pulse-height spectra were performed with no variance reduction in a pure analog mode. This method of operation is the most conservative type of Monte Carlo simulation and is required for correct use of the pulse-height tally in MCNP.

Gamma fluxes were tallied on various surfaces and with differing angular direction bins to maximize the amount of information provided. Pulse-height spectra were tallied within the active volume of the HPGe detector (excludes the 700-micron-thick outer dead layer) with and without resolution broadening. The broadening function is not quite correct for Ge, but the parameters were set to give 2.2 kilo-electron-volts (keV) at the 662-keV ^{137}Cs gamma energy, a typical resolution at this energy. The "ring detector" flux tally was used for some simulations that required spectral shapes at greater distances from a localized source, such as when the tool is positioned many feet above or below the ^{137}Cs zone. These simulations cannot produce

pulse-height spectra, however, and are useful only qualitatively to show the manner in which flux and spectral shape changes with depth for various source distributions.

Most simulations were conducted on a personal computer with a 66 megahertz (MHz) Intel 486 processor. A few simulations were performed on a faster Pentium-equipped machine, when available. Typically, the near-borehole source distribution problems were run in 4 to 5 hours, while the more difficult radially remote and uniformly distributed source problems took 12 to 48 hours to process.

2.2 Experimental Benchmarks

MCNP simulation models of borehole pulse-height spectra from the SGLS HPGe detector were benchmarked by comparing them to data taken in a borehole model designated the K Model at the Grand Junction Office (GJO) calibration facility (Steele and George 1986; Leino et al. 1994). This comparison indicates that the simulation models are accurate, in an absolute sense, from the source energy to as low as about 200 keV. Below this energy, the simulation models seem to underestimate measured count rates. These results validate the models only for a radially and axially uniform source surrounding the borehole. The model accuracy must be determined for various restricted source distributions, such as a thin annulus at the borehole wall. More benchmark data are needed and new experiments are planned for later in fiscal year 1997.

2.3 Simulated ^{137}Cs Spectra for Several Source Distributions

Three classes of ^{137}Cs source distribution are of interest in the vadose zone contaminant logging project.

The first class is the radially uniform source that is presumed in the process of calibrating contaminant photopeak response to activity concentration in picocuries per gram (pCi/g). In practice, the contaminant distribution needs to be uniform only in the first 30 or 40 cm (11.8 or 15.7 in.) of formation surrounding the borehole because relatively few gammas from larger radii reach the SGLS detector. Similarly, the source needs to extend axially, along the length of the borehole, for a distance of about 40 cm (15.7 in.) to ensure that the source meets the uniformity requirement. Whenever the contaminant distribution deviates from this uniformity condition, the reported activity concentrations, which are based on calibration with uniform source material, will be in error.

The second important class of ^{137}Cs source distribution is a contaminant localized to the region of the borehole casing. Identification of this type of source is important because such occurrences imply that contamination has migrated down the borehole, rather than through the sediment, and the reported concentrations will be incorrect. Localized contamination can occur inside the borehole, although most often it is located along the outer casing wall and in the first few centimeters of sediment extending radially from the casing wall.

The third class of ^{137}Cs source distribution of interest is a contaminant that is radially remote from the borehole. Such source distributions cannot be determined because many combinations can produce identical spectral shapes. However, as a thin annulus of source material becomes more remote radially, the spectrum eventually reaches a constant shape. Therefore, distant sources, such as pipes containing ^{137}Cs contamination that are 2 or 3 feet (61 to 91 cm) away from the borehole, will produce a unique asymptotic spectral shape.

Figure 4 shows the photon current spectrum on the curved surface of the Ge detector as simulated by MCNP for ^{137}Cs contamination uniformly distributed around a 6.25-in.-diameter borehole with a 0.313-in.-thick iron casing. Similar spectra are shown for ^{137}Cs contamination in the form of a thin layer on the inner and the outer surfaces of the casing. The spectra are normalized to the same intensity at the uncollided source energy of 662 keV. Comparison of the spectra shows that the scattered part of the spectrum contains less current when the ^{137}Cs contamination is local to the borehole than when it is uniformly distributed in the sediment (formation source).

Because the effects of detector response are not included on Figure 4, a comparison of spectra for the three source geometries shows the best contrast in spectral shape that is possible without the degrading effects of detector response. The best sensitivity-to-source location occurs just below the peak energy, in the range from about 0.5 to 0.65 million-electron-volt (MeV). For this energy range, the continuum current changes by a factor of about 10 relative to the uncollided peak current as the source changes from the inside surface of the casing to a uniform distribution. When the source is on the outside surface of the casing, the current in this energy range increases by a factor of about 2.5 as the source becomes distributed in the formation. These results indicate that the analyst should concentrate on the high-energy continuum portion of the scattered part of the ^{137}Cs spectra to determine if contamination is inside the casing. However, the entire continuum part of the spectrum, relative to the peak response, shows good sensitivity to source location.

Figure 5 presents simulated spectra for 1-cm-thick annular sources of ^{137}Cs at radial depths of 10 cm (3.9 in.) and 30 cm (11.8 in.) in the sediment. For comparison, the spectrum shape for uniformly distributed ^{137}Cs from Figure 4 is also included. All spectra are normalized to the same gamma current at 662 keV. As the ^{137}Cs contamination becomes increasingly remote from the borehole, the continuum intensity increases relative to the 662-keV uncollided intensity. Poor statistical precision precludes the simulation of spectra at radii much greater than 30 cm (11.8 in.) with present computer hardware and software capabilities. On theoretical grounds, the spectrum shape is expected to eventually stabilize as remoteness arbitrarily increases. On the basis of the trend observed in these simulations, it is expected that the asymptotic spectrum shape will be reached for sources at radii greater than about 100 cm (39.4 in.). For these large radii, the source radius cannot be inferred from the spectral shape, only that it exceeds 100 cm (39.4 in.).

Figures 4 and 5 illustrate simulated gamma current spectra incident on the surface of the SGLS detector for the nominal borehole condition but do not include the effect of detector response. The changes in spectral shape with source distribution observed in these figures are the best sensitivity to source distribution that could be expected and would be achieved only with a

Figure 4. Simulated Gamma Current Entrant on Ge Detector Surface for Three ¹³⁷Cs Sources

Figure 5. Simulated Gamma Current Entrant on Ge Detector Surface for Radially Remote Annular ¹³⁷Cs Sources

perfect detector that produced a count at the proper energy for each incident gamma. However, the HPGe detector responds by producing some counts at the source energy, with slight resolution broadening (about 2.2-keV full width, half maximum [FWHM]), and many more counts at lower energies, in the Compton continuum, from the edge energy (about 200 keV below the photopeak or source energy) to the lowest detected energies of about 60 keV. The effect of detector response is to add counts to the continuum region of the spectrum, and this causes a decrease in sensitivity to source distribution.

Figure 6 shows the simulated detector pulse-height response spectra for the incident current spectra shown on Figure 4. These spectra are also normalized to the intensity at the source energy. A comparison of spectra in Figures 4 and 6 shows that generally there is less difference in the scattered continuum portion of the spectrum, relative to the photopeak, for the various source locations with the addition of detector response. Nevertheless, for the high-energy continuum above about 600 keV, the sensitivity to source location is preserved with the addition of detector response.

These simulated spectra suggest that a ratio of the continuum counts from about 60 to 650 keV to the peak count at 662 keV can be used to determine if the ^{137}Cs contamination is local to the borehole region, radially uniform, or radially remote. If the counts in a high-energy continuum window from about 600 to 650 keV are used instead, the simulations show that the ratio should be even more sensitive to source location, though fewer counts occur at these higher energies and statistical precision decreases. These analysis concepts are presented in Section 4.1, "Methodology for Spectral Shape Factor Log Analysis."

2.4 Simulated SGLS Response to $^{90}\text{Sr}/^{90}\text{Y}$ Contamination

The bremsstrahlung spectrum measured from a $^{90}\text{Sr}/^{90}\text{Y}$ beta source distributed in the sediment surrounding the borehole was simulated with MCNP. The coupled electron-photon transport feature and the usual pulse-height tally were used for the model. The electron transport process is a time-consuming simulation in MCNP and long processing times are required to obtain statistically meaningful bremsstrahlung photon spectra. Use of the pulse-height tally in MCNP requires a purely analog simulation, with no variance reduction permitted during the transport process, and this further lengthens processing time. Consequently, the simulated bremsstrahlung pulse-height spectrum for the HPGe detector had poor statistical accuracy, even after a weekend run on a 100-MHz Pentium-equipped computer.

Figure 7 shows the electron flux spectrum for an equilibrium $^{90}\text{Sr}/^{90}\text{Y}$ beta source distributed uniformly 2 cm radially into the formation and extending ± 15.24 cm (± 6 in.) axially. The electron flux is the result of starting with the published ^{90}Sr and ^{90}Y beta yields (Browne and Firestone 1986) and transporting them with a condensed Monte Carlo approach. While the electrons transport in the formation, the bremsstrahlung flux is also tallied within this source volume as it is produced by the electrons interacting with the charge of the formation nuclei. As shown on Figure 7, the energy of bremsstrahlung photons extends to about 2 MeV. Simulation statistics are not sufficiently precise to extend this photon flux to the maximum end-point energy of 2.3 MeV that is theoretically possible. The bremsstrahlung photons are transported to the

Figure 6. Simulated SGLS Detector Pulse-Height Response for Entrant Spectra Shown on Figure 4

Figure 7. Simulated ⁹⁰Sr/⁹⁰Y Bremsstrahlung and Electron Fluxes From Sediment Sources

Figure 8. Simulated SGLS Detector Pulse-Height Response to ⁹⁰Sr/⁹⁰Y Bremsstrahlung Source

detector and a pulse-height tally is made in the detector volume, with the resulting spectrum shown on Figure 8. Although the statistical precision is poor, the integral result has a 1σ standard deviation of 14 percent. This amount of standard deviation is sufficient to at least predict the sensitivity of the tool to the presence of the parent activities from ^{90}Sr in equilibrium with ^{90}Y . When properly normalized, the simulated integral response of the detector is 0.028 count per second per picocurie per gram.

The energy of maximum count rate is about 180 keV, but the response drops rapidly with increasing energy so that most of the bremsstrahlung response occurs below about 300 keV. The count rate at 180 keV for 1,000 pCi/g of $^{90}\text{Sr}/^{90}\text{Y}$ activity is comparable to the count rate produced at this energy by the typical KUT background continuum and represents an activity approaching the practical limit of $^{90}\text{Sr}/^{90}\text{Y}$ detection. When significant continuum from ^{137}Cs is present, the minimum detectable $^{90}\text{Sr}/^{90}\text{Y}$ activity will be much larger than this 1,000-pCi/g value. If it is assumed that ^{137}Cs occurs in a formation-distributed form, then its continuum contribution from 60 to 650 keV can be removed on the basis of the 662-keV peak intensity and a stripping factor derived from the simulated spectral shape shown on Figure 6. If the KUT continuum contribution is also removed, based on their signature peaks, then the remaining continuum from 60 to 650 keV must be from $^{90}\text{Sr}/^{90}\text{Y}$. If ^{137}Cs is present but its spatial distribution is unknown, the continuum region cannot be corrected but the presence of $^{90}\text{Sr}/^{90}\text{Y}$ can still be identified because of its effect on the low-energy spectral shape.

Although the statistical precision of the total counts in the simulated pulse-height spectrum is poor, $^{90}\text{Sr}/^{90}\text{Y}$ concentrations in excess of about 1,000 pCi/g (1 nanocurie per gram) are required before a measurable signal in the low-energy continuum can be distinguished from background KUT levels. The simulated count rate from this bremsstrahlung source was 28 counts per second per nanocurie per gram for a 2-cm- (0.8-in.) thick radial source in the formation. Most of the bremsstrahlung counts occur in the energy range from 0 to 300 keV.

2.5 Discussion of Simulated Spectra

The simulation results presented in the preceding sections show that information on the spatial distribution of ^{137}Cs can be obtained from an analysis of the measured spectrum shape. A sensitive measure appears to result by calculating the ratio of the scattered continuum gammas to the uncollided 662-keV gammas; the continuum spectrum shape itself is not highly sensitive to the distribution of the ^{137}Cs source. For instance, the ratio of counts from 60 to 350 keV to counts from 350 to 650 keV varies from about 2.8 for ^{137}Cs contamination on the casing wall to about 4.5 for radially remote ^{137}Cs . On the other hand, the majority of bremsstrahlung photons from ^{90}Sr occur at energies below 350 keV so that the expected ratio is about 20 or 30, much higher than the continuum spectral shape ratio for ^{137}Cs concentration regardless of its spatial distribution. Therefore, if the measured spectra are analyzed for both the continuum-to-peak ratio and for the continuum shape ratio, it should be possible to infer the ^{137}Cs source distribution and to separate the occurrence of ^{137}Cs from $^{90}\text{Sr}/^{90}\text{Y}$.

The continuum (60 to 650 keV)-to-peak (662 keV) ratio has been designated spectral shape factor 1 (SF1) and the continuum shape ratio (counts from 60 to 350 keV divided by counts from

350 to 650 keV) has been designated SF2. Table 1 contains values for these two spectral shape factors for several source distributions. Table 1 also presents an alternative to SF1, designated SF3 (George 1996). SF3 totals only the counts in the high-energy portion of the continuum from 610 to 655 keV, which is then divided by the counts in the 662-keV peak. This measure of spectral shape is more sensitive to the source location (inside or outside the casing) than is SF1. Because of the large statistical errors in this shape factor for the radially remote source, it is difficult to determine if SF3 is more sensitive or less sensitive to source remoteness than SF1. The values in Table 1 have statistical precisions better than about 5 percent, except for the entries accompanied by precision estimates in parentheses. Although SF3 is a better indicator than SF1 of whether the source is inside or outside the casing, the measured counts are less for this high-energy window and a stronger Cs presence is required than for SF1. However, SF3 has a significant advantage over SF1 when implemented with field data because the background correction process that is due to the natural radioelements K, U, and Th is simpler and probably more accurate. The background correction is presented in Section 4.1 "Methodology for Spectral Shape Factor Log Analysis."

Table 1. ^{137}Cs Spectral Shape Factors for Several Source Distributions

^{137}Cs Source Distribution	Spectral Shape Factor		
	SF1 (60 to 650 keV/ 662-keV peak)	SF2 (60 to 350 keV/ 350 to 650 keV)	SF3 (610 to 655 keV/ 662-keV peak)
Radially Uniform in Formation	13	3.45	0.179
On Outside Surface of Casing	6.8	2.79	0.089
On Inside Surface of Casing	4.5	2.82	0.032
1 cm-Thick Annulus at 10-cm Radius	19	3.8	0.21
1-cm Thick Annulus at 30-cm Radius	37 ($\pm 25\%$)	4.2 ($\pm 25\%$)	0.71 ($\pm 50\%$)
1-cm Thick Annulus at 50-cm Radius	80 ($\pm 50\%$)	4.4 ($\pm 15\%$)	—

Shape factors were also simulated for several other source distributions and for other borehole conditions. When ^{137}Cs contamination is uniformly distributed from the outer casing wall to various radial distances into the formation, the shape factors approach their radially uniform values (row 2 in Table 1) at about 30 cm (11.8 in.). If the ^{137}Cs concentration is uniform for the first 30 cm (11.8 in.), fluctuations that occur at larger radii will not perceptibly affect measured shape factors unless concentrations are orders of magnitude greater. Figure 9 illustrates how the SF1 and SF2 values vary with the radial extent of ^{137}Cs distribution, starting with a thin coating on the outer casing surface. Most of the increases in SF1 values occur in the first 15 cm (5.9 in.)

Figure 9. Spectral Shape Factors SF1 and SF2 Values for Several Radial Extents of Formation ¹³⁷Cs

or so of radial source extent. Therefore, the measured value of SF1 is an indicator of contaminant proximity to the borehole. For example, a measured value for SF1 of 8 on Figure 9 indicates the contaminant is confined to the first 2 cm (0.8 in.) of sediment surrounding the borehole. Unfortunately, many source distributions can give the same SF1 value. For instance, it is possible for a thin contaminant coating on the outer casing wall (SF1 is equal to 6.8) to combine with a source annulus of the same count rate and a 5-cm (1.97 in.) radial extent to give a composite SF1 value of 8, identical to that for a uniform source in the first 2 cm (0.8 in.) of sediment. This example emphasizes that unique source distributions are not usually associated with the measured values of the shape factors. An exception occurs when SF1 is measured to be about 7. It is nearly certain for this instance that the contaminant is confined to the casing wall because all other source locations (except for inside the casing) produce larger values of SF1. The best approach is to conclude that the shape factors represent evidence for certain spatial source distributions. An SF1 value of 13 and an SF2 value of 3.5 are evidence of radially uniform ^{137}Cs contamination, at least for the first 30 cm (11.8 in.). The SF2 value does not vary significantly with radial source distribution but is useful in distinguishing the presence of radially remote ^{137}Cs from the presence of ^{90}Sr .

3.0 Application of Spectral Shape Factor Concepts to Logging Data

Certain conditions must be met if variations in spectral shape with the spatial distribution of ^{137}Cs are to be exploited in analyzing the SGLS vadose zone logging results at the Hanford Tank Farms for contaminant source proximity to the borehole. First, and most important, the SGLS must produce high-quality gamma-ray spectra that do not suffer distortion from the effects of excessive counting rates. Unfortunately, this condition precludes the use of spectral-shape analysis for many high-activity zones of interest that have been encountered (for example, at the SX Tank Farm) because the counting rates measured by the detector become excessive. Spectra must be free of all other contaminants, except for the bremsstrahlung-producer ^{90}Sr . The shape factor simulations presume that contaminant distribution extends sufficiently in depth so that the spectra are affected only by the radial distribution. This presumption means that the contaminant must be uniform over a depth interval of about 1.5 ft. Thinner contaminant zones will produce depth effects on the measured shape factors. Section 4.1, "Methodology for Spectral Shape-Factors Log Analysis," presents examples of the contaminant depth influence on shape factors.

The natural radioelements K, U, and Th produce a background of gammas in the low-energy continuum region that must be subtracted from the continuum energy windows before a shape analysis can be performed. The analyst can perform this correction by multiplying the measured signature peak intensities for each natural radioelement by the continuum stripping ratios determined from experiments and simulations and by then subtracting this correction from each continuum window. This computation was used to determine SF1 and SF2 values. The background correction for the narrow continuum window (610 to 655 keV) of the SF3 ratio is computed more simply. Because the continuum window for SF3 is narrow in energy, just below the 662-keV peak region, counts in a similar narrow window just above the peak are assumed to be a measure of the continuum background. The difference between the two narrow energy windows gives the high-energy continuum counts from the ^{137}Cs source distribution. Use of the K, U, and Th signature peak intensities with their inherent statistical imprecisions and the stripping ratios, which also have uncertainties, is not necessary to achieve the background correction for SF3. However, there is evidence that this simple background correction method for the SF3 continuum counts may be inadequate.

The initial application of SF1 and SF2 values to certain SGLS logging data has produced encouraging results, although there clearly are limitations to their usefulness when applied to actual logging data. One limitation concerns SF1 when evaluated for low ^{137}Cs contaminant levels. In this situation, a satisfactory KUT background correction to the continuum energy windows is difficult because the background is a large fraction of the total counts in these windows. This difficulty is caused by uncertainties in the K, U, and Th stripping ratios and by large statistical errors, generally in the U and Th signature peaks at 609 keV and 2614 keV, respectively. Because of these uncertainties, SF1 is not reliable for uniformly distributed ^{137}Cs concentrations less than about 1 pCi/g (0.6 count per second [count/s] for a uniform source distribution). This reliability threshold concentration (or count rate) is somewhat variable because the continuum counts are larger or smaller in relation to the 662-keV peak counts,

depending on source distribution. Another limitation occurs when the contaminant is present in a thin zone, 1.5 ft or less in thickness, and is bounded above and below by low contaminant concentrations. This distribution produces a shape factor anomaly that is not related to the radial extent of the contaminant and is negatively correlated with the apparent concentration anomaly. Figure 10 presents an example of computing SF1 and SF2 values in relation to depth for favorable conditions. These shape factor logs are for borehole 22-11-01 near tank BY-111. Shown for reference are two solid lines, one at 13 and the other at 3.45, that are the simulated values of SF1 and SF2, respectively, for the radially uniform ^{137}Cs distribution. Results are shown only to a depth of 30 ft because there is little or no contamination at greater depths, except at the bottom of the borehole. ^{137}Cs contamination often appears to collect at the bottoms of these cased boreholes but shape factor analysis cannot be performed on spectra from these locations because of nonstandard borehole conditions and source geometries caused by contaminant-bearing sediment material plugging the bottoms.

Figure 11, from the data report for tank BY-111 (DOE 1996a), is the combination plot for borehole 22-11-01 showing the ^{137}Cs concentration log; the K, U, and Th logs; the SGLS total gamma log; and the Tank Farms gross gamma log taken in 1989. The ^{137}Cs log clearly shows little contamination below about 30 ft. In the absence of other contaminants, an experienced analyst can identify an excess of scattered gammas at about 23 ft by comparing the ^{137}Cs log with the total gamma log. This excess of total gammas at 23 ft is an indication that the ^{137}Cs contamination may be radially remote at that depth. Many of the tank farms have considerable contamination near or at the surface. Surface contamination also is present on the log for borehole 22-11-01, with apparent ^{137}Cs concentrations between 30 and 100 pCi/g at the surface and for the first 7 ft of depth. Air above the ground surface affects the near-surface transport process and the source cannot be uniformly distributed. This nonuniform source distribution causes the reported concentration to be incorrect because the calibration factor was determined from borehole models with uniform source distributions. The spectral shape is also affected so that SF1 and SF2 values are typically larger at the surface than for radially uniform contaminant distributions at depths of about 1.5 ft or more. This is illustrated by the SF1 log on Figure 10 that shows a single high value of about 35 for the first interval logged at a depth of 0.5 ft. Below this depth, SF1 stabilizes at values between 14 and 18 to a depth of about 7 ft, indicating that ^{137}Cs contamination is distributed somewhat uniformly in the radial direction at this depth interval, at least to about 30 cm. The SF2 value is similar for this depth interval, reinforcing the interpretation of uniformly distributed ^{137}Cs contamination from the surface to about 7 ft.

The log of the ^{137}Cs peak count rate shown on Figure 10 is a valuable complement to the shape factor logs because it allows the analyst to identify intervals where the ^{137}Cs content is too low for a good shape factor analysis. Note that as the ^{137}Cs count rate drops to the 0.5 to 1 count/s range (0.8 to 1.6 pCi/g for a uniform source distribution) between 8 ft and 20 ft and below 25 ft, the SF1 and SF2 logs exhibit statistical scatter. For these intervals, some of the SF1 values are missing on the semilogarithmic statistical plot because they are negative, which is due to large error in the background correction. The point to be made is that when ^{137}Cs count rates drop below about 0.5 to 1 counts/s, any observed structure to the shape factors and their magnitudes should be interpreted with caution. The interval of elevated ^{137}Cs contamination from 22 to 25 ft

Figure 10. Shape Factor Logs for Borehole 22-11-01 Near Tank BY-111

22-11-01 Combination Plot

Figure 11. Combination Plot for Borehole 22-11-01 Near Tank BY-111 (DOE 1996a)

produces stable SF2 values between 4 and 4.5 and a peak in SF1 of about 120. These values are strong evidence that the ^{137}Cs contamination in this depth interval is radially remote from the borehole, with at least 50 cm (19.7 in.) of sediment between the contaminant and the borehole. When SF1 reaches values in the range of 100 to 200 and is accompanied by the expected rise in SF2 to about 4.5 or 5, the spectral shape is reaching its asymptotic value and the remoteness of the source cannot be determined; it could be localized at 50 cm (19.7 in.) with a certain concentration or at some greater distance with a higher concentration. SF2 provides a check on the continuum spectral shape to ensure no bremsstrahlung from ^{90}Sr is in the low-energy window. Other log examples (Figures 21 and 23) show that SF2 values can be as high as 20 or 30 when the contaminant ^{90}Sr is present.

Figure 12 presents a comparison of shape factors SF1 and SF3 for borehole 22-11-01. Recall that SF3 is a ratio of the high-energy continuum counts to the 662-keV peak counts. For zones where the ^{137}Cs count rate is 1 count/s or less (8 to 22 ft and below 25 ft), SF3 values are too large because the method for background compensation underestimates the correction. The calculation of SF3 will have to be improved if it is to be used in low-activity zones. SF1 is more correctly compensated for background and does not exhibit elevated values in these intervals. For the zone of remotely located ^{137}Cs from 22 to 25 ft, SF3 has a value of about 0.9; for the zone of uniformly distributed ^{137}Cs concentration from 2 to 7 ft, the value is 0.19. These SF3 values are consistent with those simulated for uniform and remote ^{137}Cs distributions as presented in Table 1. For the same zones, SF1 varies from about 14 to 120. In this comparison, SF1 exhibits more sensitivity to the remoteness of the source than does SF3. However, the computer modeling simulations predict that SF3 will be more sensitive than SF1 to whether a borehole-localized source is inside or outside the casing.

Figure 12. Comparison of Shape Factors SF1 and SF3 Values for Borehole 22-11-01

4.0 Spectral Shape Analyses of Tank Farms Logging Data

4.1 Methodology for Spectral Shape-Factor Log Analysis

The computer simulation of ^{137}Cs spectra for various source distributions and the rather crude simulation of the $^{90}\text{Sr}/^{90}\text{Y}$ bremsstrahlung spectrum, along with an examination of actual SGLS spectra and Tank Farms logs, provide sufficient insight into the problem of spectral shape analysis to suggest a shape-factor log analysis methodology. This initial methodology applies only to the contaminants ^{137}Cs and ^{90}Sr . However, this method may be eventually extended to include other contaminants, such as ^{60}Co , ^{125}Sb , ^{154}Eu , and possibly $^{234\text{m}}\text{Pa}$ (^{238}U). The two shape factors SF1 and SF2 are computed for specified depth intervals in a given borehole. There will be a default set of background constants with an option to recompute a background normalization on the basis of a specific analysis of a contaminant-free zone of the borehole.

The shape factor SF3, with a narrow high-energy continuum window, is still under study and may become part of the shape factor log suite when its behavior is better understood. One problem with SF3, and the main reason it is not being included in the initial methodology, is the paucity of counts in the narrow high-energy window usually necessitates use of a smoothing procedure to reduce the scatter in the SF3 log. This smoothing can cause difficulties in interpreting the log when, for example, sharp changes in the ^{137}Cs count rate occur. Also, SF3 does not appear to be as sensitive to the ^{137}Cs source distribution as SF1.

In addition to the SF1 and SF2 depth logs, the ^{137}Cs count rate log is presented, along with detector dead time as a measure of spectral quality. Another feature of the log analysis method is a cross plot of SF1 versus SF2 values. This plot includes simulated points corresponding to several ^{137}Cs source distributions; these points define a trend line that assists in interpreting measured points. The cross plot points are coded by color or symbol changes for correlation with the log plots to identify the depth zones corresponding to clustered points that, according to the trend line, are zones of remote, radially uniform, or borehole-localized ^{137}Cs contamination distribution.

4.2 Software Implementation of Shape Analysis Method

The initial implementation of the spectral shape-factor analysis concepts, many of which are still under study, is intended to provide the log analyst with a set of logs that is indicative of ^{137}Cs source distribution and that also will detect the presence of ^{90}Sr when the bremsstrahlung signal is sufficiently strong relative to the ^{137}Cs and background signals. The two shape factors SF1 and SF2 provide this information only when there is sufficient signal. For uniformly distributed or borehole-localized ^{137}Cs contamination, the 662-keV peak count rate must be 1 count/s (1.6 pCi/g for a uniform source distribution) or greater. Remotely located ^{137}Cs distributions can be detected with smaller peak count rates because the low-energy counts are enhanced. ^{90}Sr contamination can be detected from the value of SF2, in the presence of low ^{137}Cs activity, at concentrations in excess of approximately 1,000 pCi/g. This value is based on computer simulations and should be regarded only as an approximate detection limit for ^{90}Sr .

The platform for this initial implementation is the GJO computer program LOGANAL used for processing SGLS logging data. Routines were added to LOGANAL to obtain the spectral data necessary for the shape analysis, to perform the necessary computations, and to produce the desired log plots.

4.2.1 Quality Indicators

Before performing the shape factor analysis, the analyst should test the SGLS spectral data for certain indications of data quality. First, the dead time is computed from the clock time and live time as recorded on the spectra files. If the dead time exceeds about 15 percent, the spectral data for that interval are flagged as suffering from some distortion of the spectrum because of pulse pileup effects. Therefore, spectral shape information derived from such spectra is suspect.

Second, the analyst should monitor a wide energy window between 750 and 1450 keV. If this count exceeds about 5 counts/s after correction for natural background contributions from K, U and Th, another flag is added to the data. This flag indicates unexpected counts that are due to the presence of one or more contaminants other than ^{137}Cs and/or another indication of pulse pileup that is due to excessive counting rate.

A third quality indicator, not yet implemented in the software, will positively identify the presence of other contaminants. The other identified contaminants that can be expected are ^{60}Co and occasionally ^{152}Eu , ^{154}Eu , ^{125}Sb , $^{234\text{m}}\text{Pa}$ (^{238}U), and ^{235}U . If one or more of these gamma-emitting radionuclides are identified above their respective minimum detection levels (MDLs), a flag is added for each contaminant. The ^{137}Cs shape factor analysis is invalid if any of these quality flags is raised for a given depth interval.

4.2.2 Spectral Shape Analysis

Data Required—For spectra that meet the quality criteria in the previous section, an analyst can derive the following spectral information from SGLS data for the shape analysis:

<i>CL</i>	=	counts and uncertainty for 60- to 350-keV continuum window
<i>CH</i>	=	counts and uncertainty for 350- to 650-keV continuum window
<i>NH</i>	=	counts and uncertainty for 616- to 654-keV continuum window
<i>NH_{BKG}</i>	=	counts and uncertainty for 669- to 707-keV continuum window
<i>Cs</i>	=	net counts and uncertainty in 662-keV Cs photopeak
<i>K</i>	=	net counts and uncertainty in 1461-keV K peak
<i>U1</i>	=	net counts and uncertainty in 609-keV U peak
<i>U2</i>	=	net counts and uncertainty in 1765-keV U peak
<i>Th</i>	=	net counts and uncertainty in 2614-keV Th peak

Background Corrections—First the K, U, and Th background contributions to the low-energy continuum windows from 60 to 350 keV and from 35 to 650 keV are calculated. This computation uses the measured photopeak counts at 1461 keV, 609 or 1765 keV, and 2614 keV for K, U, and Th, respectively, along with stripping ratios for each continuum window derived

from a combination of computer simulations and measured spectra in calibration models enriched in K, U, and Th. Specifically, spectra from the K, U, Th, and KW models in Grand Junction were processed for the signature peak intensities as well as for counts in the continuum energy windows used for shape factor analysis. These data were used in a set of simultaneous equations that were solved for the various stripping factors for the GJO borehole condition. Correction factors were derived from simulations to account for the effect of the steel casing that is present in Hanford boreholes. The casing-corrected background stripping factors were applied to many borehole spectra, and an adjustment factor amounting to a few percent was derived that produced the best overall agreement with measured and computed continuum count logs from contaminant-free zones. Table 2 presents the adjusted stripping factors, which represent the default set for shape factor analysis. The 609-keV peak is used for the U background correction unless there is a large ^{137}Cs signal, then the 1765-keV peak is used when it is determined that this peak possesses better statistical precision. When a well-defined zone in a given monitoring borehole is free of contamination, the analyst has the option of adjusting the overall normalization of the K, U, and Th stripping ratios by requiring that the background-subtracted counts in both low-energy continuum windows be equal to 0. This option can improve the shape factor results, especially for zones of low Cs content. The continuum window background corrections are calculated as follows:

$$\begin{aligned} \text{Background for 60 to 350 keV: } BL &= 12.9(K) + 40(U1) + 90(\text{Th}) \\ \text{Background for 350 to 650 keV: } BH &= 4.1(K) + 10(U1) + 21(\text{Th}) \\ \text{Background for 60 to 650 keV: } (BL + BH) &= 17(K) + 50(U1) + 111(\text{Th}) \end{aligned}$$

Low-energy continuum stripping ratios were applied to logging data for several background zones (no contaminants); there are no apparent biases in the residuals. For some boreholes, the continuum correction is slightly high; for others, it is slightly low. The major problem is that the statistical precision of the correction, on an interval-by-interval basis, is not good, primarily because of the low intensity of the 609-keV line from the U source. The initial implementation of the background correction uses a Gaussian smoothing function that effectively averages the correction over a depth interval of about 5 ft (1.5 m). The statistical uncertainty and any systematic uncertainties in the continuum background correction establish a fundamental limitation in the ^{137}Cs concentration level at which the shape factor analysis will be successful. This level will be somewhat variable, depending on the Cs source distribution, but shape factors become unreliable for values of Cs less than 1 count/s.

Table 2. K, U, and Th Background Stripping Ratios for Shape Factor Continuum Windows

Source and Signature Energy (keV)	Stripping Ratio		
	60 to 350 keV	350 to 650 keV	60 to 650 keV
K (1461)	12.9	4.1	17
U1 (609)	40	10	50
U2 (1765)	117	29	146
Th (2614)	90	21	111

4.2.3 Shape Factor Computations

For each depth interval for which the quality criteria are satisfied, the shape factors SF1 and SF2 are computed as follows:

$$SF1 = [(CL + CH) - (BL + BH)_{AVG}] / Cs \quad (1)$$

$$SF2 = (CL - BL_{AVG}) / (CH - BH_{AVG}) \quad (2)$$

Eventually, these values for SF1 and SF2 will be accompanied by uncertainty estimates that are based on the statistical precision and any systematic error in the various quantities. To simplify the implementation in this study, SF1 is suppressed when the Cs count rate is less than 1 count/s because of a high probability of large errors. Shape factor logs presented in this report will not suppress any of the SF1 data in the interest of more effective discussions of results. SF2 is always reported, although statistical fluctuations will be apparent at low Cs levels. SF2 is always reported because of the possibility that ^{90}Sr may be present and contribute to the SF2 continuum window count, in the absence of Cs. The shape factor SF3 is similar to SF1 but uses two narrow high-energy windows NH and NH_{BKG} in place of the wide windows CL and CH :

$$SF3 = (NH - NH_{BKG}) / Cs \quad (3)$$

SF3 is included in the log analysis software as an optional shape factor. For this initial implementation, the analyst can plot SF3 in situations where it may be a superior indicator of Cs location. SF3 may suffer from a systematic error at low Cs count rates because of the imperfect background compensation represented by the simple difference $NH - NH_{BKG}$.

4.2.4 Spectral Shape Factor Log Examples From Tank Farms Boreholes

Figures in subsequent sections represent examples of shape factor logs that illustrate the application of the methodology to SGLS logging data from Hanford Tank Farms monitoring boreholes. The normal presentation for the initial implementation of the software consists of depth logs for SF1, SF2, the ^{137}Cs peak count rate, and detector dead time. This normal presentation is linear for the depth scale and logarithmic for the three log parameters. Also shown, for reference, are solid lines corresponding to the simulated values of 13 for SF1 and 3.45 for SF2 for radially uniform ^{137}Cs . These logs are usually accompanied by a cross plot of SF1 and SF2 values for selected depth intervals. When possible, a contaminant-free section of the borehole is used to make adjustments in the background stripping ratios given in Table 2 to improve the K, U, and Th background correction. This process has often led to an approximately 10-percent increase in the stripping ratios, indicating that the values in Table 2 may be too small.

4.2.5 Radially Uniform ^{137}Cs Distributions

In the borehole data processed thus far for the shape factor logs, ^{137}Cs contamination appears to be uniformly distributed in the radial direction in only a few depth intervals. For uniformly

distributed ^{137}Cs , the values of SF1 and SF2 are predicted to be 13 and 3.45, respectively (see Table 1).

For the limited number of boreholes that have been studied for shape factors thus far, SF1 and SF2 indicate the presence of uniformly distributed ^{137}Cs in some depth intervals. One example is shown on Figure 13 for borehole 22-12-03 near tank BY-112 (DOE 1996b): ^{137}Cs contamination appears to be radially uniform from 9 to 18 ft and from 23 to 40 ft. The ^{137}Cs peak count rate from 19 to 23 ft is too small (less than about 1 count/s) to determine reliable SF1 values or the source distribution, but the distribution could be uniform. It is unlikely that the SF1 anomaly from 20 to 23 ft is due to remote ^{137}Cs contamination, rather it is an example of the negative correlation that occurs between SF1 and Cs when there are large changes in Cs distribution over short depth intervals, or, alternatively, it could be due to slight undercorrection of the continuum window for the KUT background. For depths above 9 ft, the spectral shape logs are not calculable because quality indicators show the presence of spectral distortion from pulse pileup as well as the presence of the additional contaminants ^{60}Co and ^{154}Eu . In particular, at depths from 3 to 7 ft, dead time exceeds the value of 15 percent that is considered the upper limit for good quality spectra. In summary, this borehole exhibits uniformly distributed contamination for a considerable fraction of its total depth. Below 40 ft, the ^{137}Cs peak count rate is less than the 1 count/s required to perform the shape factor analysis.

Another example of radially uniform ^{137}Cs is shown on Figure 10 for borehole 22-11-01 near tank BY-111. Because little or no ^{137}Cs contamination is indicated below a depth of 30 ft, the shape factor logs are shown only from the surface to 30 ft. From about 1 to 7 ft, SF1 values range from about 14 to 17 and the SF2 value is quite stable at about 3.5, indicating uniformly distributed ^{137}Cs . Below this depth, the Cs content drops below 1 count/s and the statistical fluctuation in SF1 is quite large, making interpretation difficult. From 23 to 25 ft, the Cs source appears to be remote from the borehole because of elevated values for both SF1 and SF2.

4.2.6 ^{137}Cs Distributions Local to Boreholes

Shape factor logs for borehole 41-09-04 near tank SX-109 (DOE 1995d) show strong evidence that the ^{137}Cs contamination is local to the borehole casing region for depths from 3 to 50 ft. Figure 14 shows that both SF1 and SF2 are well below the values expected for uniformly distributed ^{137}Cs concentrations. Because the quality indicators are satisfied for this depth range and because the Cs count rate is sufficiently high, the precision of SF1 and SF2 is good for all depths. Dead time exceeds 15 percent for depths from the surface to 3 ft and, therefore, shape factors are not valid for this depth range. A high contamination zone, starting at about 57 ft, causes both SF1 and SF2 to gradually increase as this zone is approached from above, starting at about 42 ft. Above 42 ft, the shape factors are smaller than expected for uniformly distributed Cs and are diagnostic of contaminant localized to the borehole region. Values of SF1 range from about 8 at the 5-ft depth to a low of about 6 from 20 to 40 ft. Values of SF2 are also less than expected for uniformly distributed Cs. In the interval from 20 to 40 ft, SF2 is about 2.8. Of the logs examined so far, these results provide the best evidence for Cs at or close to the outer wall of the borehole casing. Nearer the surface where SF1 and SF2 are somewhat larger, the Cs

Figure 13. Shape Factor Logs for Borehole 22-12-03 Near Tank BY-112

Figure 14. Shape Factor Logs for Borehole 41-09-04 Near Tank SX-109

distribution may extend as far as about 4 cm (1.6 in.) into the surrounding sediment (refer to Figure 9 for the simulated dependence of spectral shape on radial source extent).

Figure 15 presents shape factor logs for borehole 41-12-02 near tank SX-112 (DOE 1995f). For the entire depth range from near the surface to about 50 ft, both SF1 and SF2 have values indicative of Cs local to the borehole region, extending no more than about 6 cm (2.4 in.) radially from the casing wall into the sediment. Below 50 ft, the streaming effect of a hot zone at lower depths complicates the interpretation of the shape factors; shape factors are not shown for these lower depths in this borehole. From about 35 to 50 ft, SF1 values between 7 and 8 indicate the Cs contamination is at the outer casing wall with little radial extent into the sediment. Figure 16 illustrates these source locations on a cross plot that shows the simulated points for various radial source extents: inner casing wall, outer casing wall, first 2 cm (0.8 in.) of sediment, first 6 cm (2.4 in.) of sediment, and radially uniform in the sediment. The points are joined by a line to indicate the trend for SF1 and SF2 with increasing radial source extent. These data are in the range of values for the source at the casing wall and the source extending to about 6 cm into the sediment. In a few depth intervals (27 to 29 ft and 34 to 35 ft), SF1 and SF2 approach the values of 13 and 3.45, respectively, for uniformly distributed Cs.

4.2.7 Remote and Local ^{137}Cs Distributions in a Single Borehole

Logs for borehole 41-02-02 located near tank SX-102 (DOE 1995b) illustrate the use of shape factor analysis to identify remote and local ^{137}Cs distributions. Figure 17 presents the shape factor log plot for the depth range from 0 to 75 ft in borehole 41-02-02. The published results for SGLS logging around this tank show little Cs contamination below a depth of 75 ft, except near the bottom of the well at 140 ft (DOE 1995b). That report also suggests that the contamination from 47 to 54 ft may contain ^{90}Sr as well as ^{137}Cs . However, the shape factor logs on Figure 17 show that the enhancement in scattered continuum in relation to the Cs peak count rate, as manifested in a SF1 value of about 100 and the accompanying increase in SF2 to a value of about 5, is evidence for the presence of radially remote ^{137}Cs . If ^{90}Sr were present, the value of SF2 would have to be larger than 5. This shape factor analysis illustrates an important use of the technique to distinguish between the presence of ^{90}Sr and radially remote ^{137}Cs contamination.

From 7 to 12 ft, the Cs count rate is substantially above the 1 count/s minimum for reliable values of SF1. The values of about 8 and 3.2 for SF1 and SF2, respectively, indicate that Cs contamination is not radially uniform but is concentrated in the near-borehole region, probably in the first 2 to 4 cm (0.8 to 1.6 in.) of surrounding sediment. Another zone where Cs contamination is local to the borehole region occurs from 32 to 37 ft. Cs appears to be uniformly distributed radially from 3 to 6 ft. The Cs peak count rate is too low to obtain reliable SF1 values from 14 to 16 ft, 23 to 27 ft, and 66 to 68 ft. It is possible that the thin zone anomaly at 70 ft is uniform radially because SF1 approaches the value of 13 expected for axially thick, radially uniform ^{137}Cs .

Figure 18 presents the cross plot of SF1 compared with SF2 values for borehole 41-02-02 near tank SX-102. The clustering of points between values for Cs on the casing wall and radially uniform Cs is readily apparent, along with the values from 47 to 52 ft that correspond to radially

Figure 15. Shape Factor Logs for Borehole 41-12-02 Near Tank SX-112

Figure 16. Cross Plot for SF1 Versus SF2 Values for Borehole 41-12-02 Near Tank SX-112

Figure 17. Shape Factor Logs for Borehole 41-02-02 Near Tank SX-102

Figure 18. Cross Plot for SF1 Versus SF2 Values for Borehole 41-02-02 Near Tank SX-102

remote Cs. The trend line is based on computer simulations. SF2 values seem to be slightly higher than expected for a remote Cs concentration. These higher values could be due to the presence of a slight ^{90}Sr component or, more likely, to errors both in the simulations and in the background-corrected shape factors.

Figure 19 shows the shape factor logs for borehole 41-02-08, also near tank SX-102 (DOE 1995b). These results are similar to those for borehole 41-02-02. A radially remote Cs source is also detected in this borehole from about 45 to 52 ft. Shape factor logs for the depth interval from 3 to 17 ft show a Cs occurrence that is somewhat localized to the borehole region; this localization is more obvious in the cross plot shown on Figure 20. There is a negative correlation effect between the SF1 value and the Cs count rate that may be due to the rather strong variation of concentration with depth. There is no evidence of ^{90}Sr contamination from 45 to 52 ft, although the possibility of its occurrence was mentioned in DOE (1995b). This interpretation is based on the clustering of points near the 50-cm (19.7-in.) remote point on Figure 20. The presence of ^{90}Sr would cause the cluster to move well above this point on the trend line. Cs count rates are too small for reliable SF1 values from 18 to 44 ft.

4.2.8 ^{90}Sr Identification in Presence of ^{137}Cs

One of the few positive identifications of ^{90}Sr contamination occurs near the bottom of borehole 41-08-11 located near tank SX-108 (DOE 1995c). This identification is based on anomalously high values of SF2 from about 65 to 74 ft. Figure 21 presents the shape factor logs for this borehole. A gap in data from 48 to 62 ft is the result of excessive count rates and pulse pileup that severely distort the measured spectra. This zone contains so much gamma activity that the spectral shape information, especially the value of SF1, above and below the 48- to 62-ft interval is affected by gammas streaming to all depths. Under such conditions, SF1 cannot be used to determine source distribution for that depth. SF2 is less affected by the hot zone and is observed to change more slowly with vertical distance than SF1.

^{90}Sr contamination is identified below the hot zone, between about 65 and 73 ft. For this depth range, SF2 is relatively constant with values ranging from about 14 to 17, far in excess of what is expected for the spectral shape from any possible ^{137}Cs source distribution. Figure 22 shows a cross plot of SF1 and SF2 values for borehole 41-08-11. The plot presents "circles" as the shape factors from 65 to 74 ft, just below the zone of high activity. This cluster represents SF1 values ranging from about 40 to 600 and maximum SF2 values of about 17. The trend line for the occurrence of ^{137}Cs at varying radial distributions ranging from local to the borehole to radially remote at 50 cm (19.7 in.) is shown for comparison. The squares correspond to all other depth intervals for this borehole. SF1 values for points close to the hot zone are no doubt inflated by the streaming effect and, hence, are larger than expected for a radially remote Cs source. However, the SF2 value should be diagnostic of remote Cs or of ^{90}Sr because SF2 varies slowly with distance from a hot zone. The shape factor cluster (circles) far above the trend line represents strong evidence for the presence of ^{90}Sr , along with remotely located ^{137}Cs contamination.

Figure 19. Shape Factor Logs for Borehole 41-02-08 Near Tank SX-102

Figure 20. Cross Plot for SF1 Versus SF2 Values for Borehole 41-02-08 Near Tank SX-102

Figure 21. Shape Factor Logs for Borehole 41-08-11 Near Tank SX-108

Figure 22. Cross Plot for SF1 Versus SF2 Values for Borehole 41-08-11 Near Tank SX-108

4.2.9 ^{90}Sr Identification in the Absence of ^{137}Cs

Figure 23 is a linear plot of SF2 and Cs count rate versus depth near the bottom of borehole 41-11-10 located near tank SX-111 (DOE 1995e). Near the bottom of the well, between 115 and 120 ft, there is a well-defined peak in the value of SF2 where it rises to a value of about 48. There is no evidence from the published SGLS total gamma log or the ^{137}Cs log (DOE 1995e) of contamination or enhanced scattered radiation at this depth. The magnitude of this SF2 anomaly, representing an enhancement in low-energy counts, is uncertain because of the difficulty in performing the background correction. This anomaly exhibits larger SF2 values than observed from 65 to 74 ft in borehole 41-08-11 where ^{137}Cs was also present (Figure 22). This may indicate the presence of a more pure ^{90}Sr source at this depth in borehole 41-11-10. The spectra were examined at a depth of 117 ft where SF2 has its maximum value of 48. When background from K, U, and Th is subtracted, the residual continuum is concentrated in the energies below about 200 keV. This is further evidence for the presence of ^{90}Sr . This result suggests that, in the future, the sensitivity of SF2 to the presence of ^{90}Sr could be enhanced by changing the boundary between the energy windows used in the continuum count ratio from the presently used 350 keV to a lower energy of about 200 keV. If the net continuum counts are summed and the simulated ^{90}Sr sensitivity is used to derive a calibration factor, the ^{90}Sr content is estimated to be about 8,000 pCi/g at 117 ft. Of course, this is only an approximate concentration because the simulated ^{90}Sr spectra have large statistical errors. In addition, the SF2 anomaly contains inaccuracy that is due to uncertainty in the background correction.

Figure 24 is a logarithmic plot of the same quantities from the surface to a depth of 125 ft. SF2 values appear to be elevated for most depths. The SF2 anomaly at 117 ft is the same one shown on Figure 23 and is attributed to the presence of ^{90}Sr contamination. There are three depth zones where the quality indicators (percent dead time, in particular) show the spectral shapes are not usable because of spectral distortion: 63 to 73 ft, 76 to 79 ft, and 82 to 85 ft. For all other depths, the spectral quality is good and the only contaminants expected are ^{137}Cs and/or ^{90}Sr . SF1 is not plotted because the Cs count rates either do not meet the minimum value of 1 count/s or the quality indicator value for spectral distortion is exceeded. SF2 values are high, ranging from 4 to 6 from 0 to 60 ft and are statistically fairly stable even though the Cs count rate is low. Except for values from 55 to 60 ft near the hot zone, which are affected by streaming of scattered gammas, the elevated SF2 values are evidence either of low-intensity remotely located ^{137}Cs or of ^{90}Sr contamination. It is also possible that the background correction is incomplete, producing a false SF2 anomaly at these depths.

Figure 23. Shape Factor SF2 and ^{137}Cs Count Rate Logs for 85- to 125-ft Interval in Borehole 41-11-10 Near Tank SX-111

Figure 24. Shape Factor SF2 and ^{137}Cs Count Rate Logs for Entire Length of Borehole 41-11-10

5.0 Additional Effects of Contaminant Source Distribution

5.1 Effect of Tool Position Within a Borehole on Measured Spectra

Simulations were performed for a ^{137}Cs source layer on the inside wall of the casing and for the centered and eccentric tool positions. The borehole inside diameter is 6.25 in. and the tool outside diameter is 3.375 in. The difference in ^{137}Cs peak intensity is about 5 percent for these two positions, with the eccentric position having the highest response. No such position dependence is expected for the formation-distributed source and the effect should be less than 5 percent for the source on the outer casing wall. Wilson and Conaway (1993) reported the dependence of tool response on borehole position for a gamma source localized to the borehole region for similar conditions but for potassium and radon-daughter gamma rays.

Simulations were conducted for a line source of ^{137}Cs and for several azimuthal orientations relative to the eccentric SGLS tool (Stromswold 1995). Figure 25 shows the effect of this orientation on tool response for the 662-keV intensity for line sources both inside and outside the casing. Table 3 presents the numerical data plotted on Figure 25. As expected, the effect is largest for the inside casing source and varies by a factor of 1.76/0.68 that is equal to 2.6 from the 0° orientation to the 180° orientation. When the gamma source is not symmetric about the borehole axis, a large tool position effect occurs that suggests the tool should be operated in a centered mode for such source conditions to eliminate the position effect.

Table 3. SGLS Tool Response to a Line Source for Several Azimuthal Positions of an Eccentered Tool Within a Borehole

Eccentered Tool Position Relative to Line Source (degrees)	Tool Response Normed to Centered Response for Line Source Inside Casing	Tool Response Normed to Centered Response for Line Source Outside Casing
0	1.76	1.59
90	0.91	0.90
180	0.68	0.72

5.2 Spectral Shape Dependence on Axial Depth From a Vertically Thin Contaminant Zone

Spectra were simulated for several depth locations within the borehole, either above or below a 1-ft (30.5-cm)-thick horizontal contaminant zone. Three horizontal distributions were included: uniform in the formation, thin coating on inner casing wall, and thin coating on outer casing wall. The detectors were vertically displaced from the midplane of the contaminant zone by distances of 0, 50, 100, 200, 400, 800, and 1,200 cm (0, 1.64, 3.28, 6.57, 13.1, 26.3, and 39.4 ft). No tool or detector was used in this model. For the MCNP simulation model, an idealized ring detector was placed at each depth within the cased borehole. This tally provides good statistical

Figure 25. Effect of Tool Position Within the Borehole on Response to Line Sources of ^{137}Cs

precision at large distances from the source but can produce misleading results when particle tracking takes place at large distances from the detector ring. Results for distances at 400 cm (13.1 ft) and greater should be used with caution until verified with experiments. Better simulations for this geometry are expected when discrete-ordinates transport models are installed and tested on a recently acquired workstation.

Figure 26 presents the flux spectra incident on each detector location for the source on the inside casing wall with an axial extent ± 15.24 cm (6 in.). The spectra are normalized to a common value at 662 keV. These spectra show that the 662-keV peak persists at all depths, even at 1,200 cm (39 ft) from the source. They also predict the rate at which this peak intensity falls off with distance from the source. For example, as the detector moves from 200 to 800 cm (6.57 to 26.3 ft) from the source, the peak intensity drops by about a factor of 16, close to a $1/z^2$ dependence. These spectra also show that the scattered continuum portion of the spectrum becomes relatively less intense, in comparison with the 662-keV peak, with increasing distance from the source plane. This decrease in intensity means the peak should become more intense relative to the continuum as the detector moves away from a source inside the borehole. Therefore, the SF1 value will decrease with increasing axial distance from the source plane. In the SGLS logs from Tank Farms boreholes examined thus far, the only example of this so-called "shine" from contamination inside the casing was observed in logs for borehole 41-08-07 near tank SX-108 (DOE 1995c). Smoothly decreasing values of SF1 and Cs peak count rate with increasing distance from the zone of contamination represent clear signatures for contamination inside the casing. However, the indicators probably will be useful only for high contamination levels and when the remainder of the borehole does not contain significant contamination. A more direct indication of the presence of ^{137}Cs inside the borehole are the x rays at 32 and 36 keV from the daughter $^{137\text{m}}\text{Ba}$ (see low energy end of spectra on Figure 26). However, the x-ray energy is too low to be measured by the SGLS because of its thick housing and because of the detector dead layer. A different detector and housing would be able to measure these x rays as an inside-the-casing indicator of Cs contamination.

Figure 27 presents a similar set of spectral plots for the source on the outside wall of the casing, again with axial extent ± 15.24 cm (6 in.). For this source location, the peak intensity attenuates quickly with depth and disappears about 200 cm (6.57 ft) from the source plane. This behavior is logical because the uncollided source gammas must pass through ever-increasing thicknesses of iron as the distance from the source plane increases, substantiating that the continuum becomes dominant for larger distances. This result predicts that a continuum flux will persist for large distances from the source plane, that the SF1 value will rise as the axial distance from the source plane increases, and that this trend will persist in the absence of Cs sources at the local depth of the tool. In practice, however, the continuum from the distant hot zone combines with the nonzero local Cs count rate to produce a decreasing SF1 value as the distance from the hot zone increases. The SF1 log, therefore, will rise with distance from the hot zone for the first 200 cm (6.57 ft) or so and then will begin to fall as the axial distance from the hot zone increases. This effect appears in several SGLS logs that involve hot zones of Cs concentration and explains the negative correlation between the SF1 value and the Cs count rate observed in many logs where the Cs count rate is fairly high and shows large variation with depth.

Figure 26. Gamma Flux Dependence on Depth From Thin Vertical Zone of ^{137}Cs Inside the Casing

Figure 27. Gamma Flux Dependence on Depth From Thin Zone of ^{137}Cs on Outer Casing Wall

An example of the spectral behavior with increasing depth from a hot zone outside the casing wall is the logs for borehole 41-09-03 near tank SX-109 (DOE 1995d). Figure 28 presents the combination log plot for this borehole. A zone of strong gamma activity is apparent from 60 to 70 ft, where the detector was reported to be saturated (DOE 1995d). If the spectra are examined just above this zone, the 662-keV peak intensity (represented by the ^{137}Cs concentration log in the left track of Figure 28) is observed to fall off rapidly to negligible values after about 5 ft (91.44 cm), whereas the continuum associated with ^{137}Cs contamination from 60 to 650 keV is observed to persist for about 30 ft (914 cm) above the hot zone. This behavior is manifested in the logs by a persistent total gamma response above the hot zone while the apparent ^{137}Cs concentration, in the absence of a local Cs source, falls to values less than 1 pCi/g by about 3 ft (152 cm) above the hot zone. When these spectra are examined, the total gamma response is clearly from counts in the low-energy scattered continuum, as predicted by the simulations. The presence of an enhanced continuum component from ^{137}Cs indicates a remote source location. If this remoteness is due to a large depth difference between the detector and the depth-localized formation source, there will be a smooth decrease in the continuum with distance from the hot zone. If the remoteness is due to a source in the formation that is distant from the borehole at all depths, the enhanced continuum will have a depth dependence similar to that for other normal formation gamma logs, rather than the smoothly changing values observed in the logs for borehole 41-09-03. Therefore, the two types of "remoteness" can be distinguished from the character of the logs.

Figure 29 presents the corresponding set of spectra for the source uniformly distributed in the formation with an axial extent of ± 30.48 cm (12 in.). The trends are similar to those for the source on the outside casing wall, as shown on Figure 27, except the continuum is more intense relative to the peak for the shorter depth increments from 0 to 100 cm (3.28 ft), as expected. The peak intensity disappears quickly with depth from the source plane. As with a depth-localized source on the outer casing wall, the SF1 value will at first increase with distance from the hot zone but then will start to decrease with greater distances because of the combined effect of remotely located continuum gammas and local 662-keV peak gammas. This effect is a probable explanation for the negative correlation observed between the SF1 value and Cs when peak count rates are strongly varying over short depth intervals.

41-09-03 Combination Plot

Figure 28. Combination Log Plot for Borehole 41-09-03 Near Tank SX-109 (DOE 1995d)

Figure 29. Gamma Flux Dependence on Depth From Thin Zone of ^{137}Cs Uniformly Distributed in Sediment

6.0 Summary

Spectral shape analysis techniques were developed for SGLS data acquired in the boreholes that surround the single-shell waste storage tanks at the Hanford Site. These techniques enable a log analyst to determine, in many instances, if ^{137}Cs contamination is uniformly distributed in the sediment surrounding the cased boreholes, is localized in the vicinity of the borehole casing, or is radially remote from the borehole. These analysis methods were also used to identify the presence of ^{90}Sr from its bremsstrahlung signature in the low-energy portion of the gamma spectra. Interpretation methods for measured spectra are based on computer modeling simulations that predict spectral shapes for the several source distributions and on calculations of spectral shape factors that, when used collaboratively, determine the Cs distribution and the presence of ^{90}Sr .

Simulations show that a large value for the continuum-to-peak spectral shape ratio SF1, larger than the value of about 13 expected when adjacent to a formation-distributed source, is indicative of a source location that is axially and/or radially removed from the detector location and should be flagged as leading to erroneous ^{137}Cs concentrations for that location. If the spectral shape ratio is significantly less than 13, the source is local to the borehole (either inside or outside the casing). If the ratio is less than the value of about 4.5 simulated for a source on the inside wall of the casing, the contamination is inside the borehole and at a different depth than the detector. All of these cases suggest that the logged concentrations for ^{137}Cs be flagged because the source conditions deviate from the calibration condition of a uniformly distributed formation source. Simulations predict that spectral shape ratio SF2 is 3.45 for uniformly distributed ^{137}Cs and rises slowly to about 5 for radially remote ^{137}Cs . When ^{90}Sr is present, the SF2 value rises sharply to values in excess of about 20. SF1 and SF2 values used together provide a means of distinguishing between a radially remote ^{137}Cs source and the bremsstrahlung from ^{90}Sr .

Shape-factor analysis techniques were tested on a number of SGLS logs, primarily from boreholes surrounding the waste storage tanks in the SX Farm. The analysis includes the presentation with depth of two shape factors: SF1 that is a measure of the scattered continuum gammas relative to the 662-keV peak gammas and SF2 that is a measure of the shape of the continuum gamma distribution. SF3, an optional third shape factor, is a measure of the high-energy scattered continuum gammas relative to the 662-keV peak. SF3 may be preferable to SF1 to distinguish between Cs contamination on the inside and the outside surfaces of the casing and when it is difficult to correct for the KUT background in the continuum portion of the gamma spectra. The nominal shape factor log presentation includes SF1 and SF2 values and the 662-keV peak count rate. A cross plot of SF1 and SF2 values is useful to separate the occurrence of a radially remote ^{137}Cs source from ^{90}Sr contamination and to illustrate the clustering of data around values that simulations predict correspond to the various possible ^{137}Cs source distributions.

Experimental verification of the simulations is needed. Experiments in preparation will use point sources of ^{137}Cs and ^{90}Sr at various radial distances from a borehole casing in a sand-filled tank.

These experiments will provide important benchmark spectra for validation of the simulation models.

Enhancements to the computer simulation capability in progress include the acquisition of a fast Pentium Pro workstation and the implementation of new software to improve the simulation of bremsstrahlung spectra from ^{90}Sr and of spectra from remote sources of ^{137}Cs . The database of simulated spectra will be expanded to include the gamma-emitting radionuclides ^{60}Co , ^{152}Eu , ^{154}Eu , ^{125}Sb , and $^{234\text{m}}\text{Pa}$ (^{238}U).

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