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High-Performance Gamma Measurements of Equipment Retrieved from Hanford High-Level Nuclear Waste Tanks

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High Performance Gamma Spectroscopy Measurements of Equipment Retrieved From Hanford High Level Nuclear Waste Tanks[1]. G. L. Troyer, K. E. Hillesand*, S. G. Goodwin**, S. F. Kessler*, E. W. Killian**, D. Legare***, J. V. Nelson, Jr.*, R. F. Richard*, and E. M. Nordquist. Numatec Hanford Co., Richland, WA, 99352, USA.

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Abstract

The cleanup of high level defense nuclear waste at the Hanford site presents several progressive challenges. Among these is the removal and disposal of various components from buried active waste tanks to allow new equipment insertion or hazards mitigation. A unique automated retrieval system at the tank provides for retrieval, high pressure washing, inventory measurement, and containment for disposal. Key to the inventory measurement is a three detector HPGe high performance gamma spectroscopy system capable of recovering data at up to ninety per cent saturation(200,000 counts per second). Data recovery is based on a unique embedded electronic pulser and specialized software to report the inventory. Each of the detectors have different shielding specified through Monte Carlo simulation with the MCNP program. This shielding provides performance over a dynamic range of eight orders of magnitude. System description, calibration issues and operational experiences are discussed.

Introduction

The Hanford site has a large quantity of highly radioactive chemical wastes stored in underground storage tanks. The waste is predominantly supersaturated aqueous solutions of sodium nitrate, aluminum nitrate, and sodium hydroxide with dose levels of up to 7 Gy/hr (700 R/hr). An example configuration of the tanks is shown in Figure 1. The wastes are the results of over 50 years of nuclear fuels reprocessing stemming from the World War II Manhatten Project. The current mission is to clean up the Hanford site by stabilizing and reprocessing the chemical inventory.

As shown in Figure 1, the tanks contain various sensor or equipment systems. In particular, liquid level devices and thermocouple trees are found in multiple quantities. These are deployed from above ground through tank top pipe risers or well heads. The tanks have up to 3.8×10^6 liter (1 million gal.) capacity, and the largest are 22 m (75 ft) in diameter and 12 m (40 ft) high. The

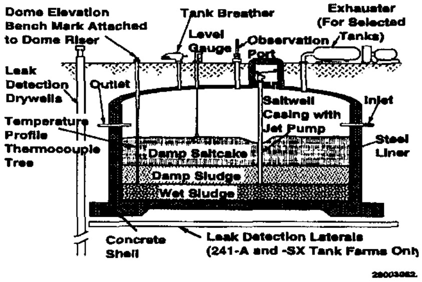

Figure 1. Example Hanford site high level nuclear waste storage tank.

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[1] Work performed for the USDOE under contract DE-AC06-87RL10930.

access risers are small, typically 10 to 20 cm (4 to 8 in) in diameter. In order to install new equipment or prepare for new operations, the existing equipment must be removed.

The removal of the equipment presents several challenges. The integrity of the equipment is suspect and where hollow cavities exist, invasion of waste material is expected. In addition, the saturated nature of the wastes has caused significant corrosion and radioactive salt cake formation on the equipment. As equipment is removed, the advantage of soil overburden shielding of radiation is lost resulting in potential significant radiation exposure to workers. Thus, a consolidated effort is made to reduce the radioactive materials such that the removed equipment can be safely handled and disposed.

System Design Criteria

Primary criteria for these operations are the provision of equipment capable of removing, cleaning and contamination reduction, transport, and storage or disposal, of contaminated equipment and fixtures as required to support operations (WHC, 1995). Additional criteria include the measurement of activity level of the equipment as it is withdrawn from the tank and the removal of residual waste liquid and solids. Use of any chemical cleaning agents other than water is precluded so as not to change tank contents category from ‘mixed waste’ to ‘listed waste’ (WAC 173-303-080). During component removal and handling, tank dome loads are limited precluding direct positioning of cranes or other heavy equipment (Bergmann 1988). Finally, working components must provide and maintain containment of radioactive solutions and vapors, including provisions for containment of any solutions which may leak as it is being removed from the waste tank. Any leak of solution to ground surfaces is unacceptable.

The system was designed to include a hoisting crane, a high pressure wash system, primary and secondary containment, remote radiation measurements and system control. A conceptual layout of the system is shown in Figure 2. Once set, all operations are controlled remotely up to 61 m (200 ft) from the removal point. The crane provides standoff outside the tank dome perimeter and hoisting and transport of the retrieved equipment to overpack containment. The wash system provides up to 21 MPa (3000 psi) with up to 66°C (150°F) water. Primary containment is with an automatically deployed and flexible accordion heavy gauge plastic bag. Secondary or outer containment is a steel shipping cask. The flexible receiver includes video cameras for control monitoring, radiation detectors, and hydraulic bag cutters and clamps.

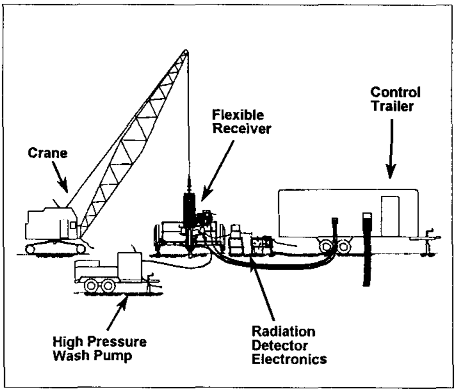

Figure 2. Conceptual view of flexible receiver system.

Radiation Measurement System

The Flexible Receiver Radiation Detection System (FRRDS) provides a high capacity measurement system for gamma photon spectroscopy. It is based on state of the art high purity solid state germanium (HPGe) gamma detectors coupled to high speed and self correcting electronics. The detector system provides precise sorting of the gamma photons such that the source radionuclide can be identified and the quantity recorded. The design configuration and modeling are described here.

Configuration

Three low efficiency (15% relative to NaI(Tl) 3x3) HPGe detectors were equally positioned on a common diameter outside the FRRDS body spool piece. Position mounts were provided with 5 cm (2 in.) of lead shielding on the top and sides and 10 cm (4 in.) on the bottom of each detector. The space between the end of the detector and the spool piece included a different shield thickness for each detector based on model calculations described below. This provided a dynamic range approximating five orders of magnitude ($1x10^5$) beyond normal counting range.

A novel electronic pulser system was used to insure that the radiation system provided measurement stability. This system (Killian, 1992, 1988) periodically injects a precision electronic pulse into the detector electronic circuits. As activity or counting rate increases, pulses are lost and changes in resolution and gain can occur. The character and amount of loss of pulser events over expected can be applied as correction factors to normal gamma spectroscopy data. This allows the system to be operated at high data rates while still obtaining useful information.

The FRRDS was configured to have the detectors and initial signal conversion and storage equipment located at the well head. An ethernet control and data link was provided from the well head to a remote control trailer which housed the spectroscopy reduction system. All operations except pre-job startup can be performed from the trailer at distances up to 61 m (200 ft).

An encoder was used to transmit crane cable position to the control trailer on demand of the spectroscopy system. A post processing program was used to merge the crane position data to the radiation information based on system clock time. The spectroscopy system acquired data at one minute intervals. The crane data were requested on a 5 second interval. At a retrieval rate of a foot per minute, the spectral position match up is within 5 cm (2 in).

Shielding Analyses and Calculational Models

To insure proper operation of the FRRDS, physics modeling of the effect of materials, removed object geometries, and detector characteristics on radiation intensities was necessary. It was anticipated that the radiation intensity or photon flux could exhibit a wide range. The expected range was from minimal background to totally swamping the detectors. Therefore, detailed analysis for a graded shielding system was performed to:

1. design a detector collimator arrangement that provided sufficient range and count rate overlap for all conceivable radiation levels,

2. determine if the detector housing design provided sufficient shielding, and

3. calculate, for each detector, geometry correction factors for the contamination levels recorded by the FRRDS data acquisition system.

Two radionuclide source terms (Hetzer 1995) were considered in the shielding analyses. Both were results of historical sampling of the tank contents. The first was based on radionuclide concentrations measured for the liquid phase of the waste products in Tank 101-AZ. The second was based on radionuclide concentrations measured for the sludge at the bottom of the tank. Radionuclide concentrations were converted to photon source rates as a function of energy using the ORIGEN-2 program (Croff 1980).

Photon flux calculations were performed using the Monte Carlo program, MCNP (Breismeister 1993). The important features of the FRRDS hardware, the thermocouple instrument trees (IT), and the geometry of the tank riser were appropriately represented in the MCNP models. Although their structure varied with type and elevation, the ITs were modeled as a uniform pipe with an average outer diameter of 12.6 cm (5 in) and a thickness of 0.6 cm (.25 in). Omission of IT features such as joints or strengthening fins was judged to have a negligible impact on the results relative to other uncertainties.

A range of contamination levels remaining on the ITs was analyzed with MCNP. This extended from very low residuals to an extreme case where the IT pipe was assumed to be filled with tank waste and heavily coated on the outside with encrusted waste material. This wide range was considered to insure that the detector shielding was configured to adequately assess all potential contamination levels.

As in most MCNP calculations, the incoming photon currents at the surfaces of the simulated germanium detector were tallied. Photons entering the detector region were terminated to insure that only incoming currents were tallied and that a photon would not be scored more than once. The peak efficiency curve measured for the FRRDS (unshielded) was used to determine the factors necessary to convert a computed photon current to a detector count rate. These energy-dependent factors were included in the MCNP input data as tally multipliers.

For direct comparison with measured efficiencies, MCNP was also used to compute the peak germanium detector efficiency as a function of energy for comparison with measured data. The MCNP-calculated efficiencies averaged about 25% higher than the measured values, with the exception of the lowest photon energy computed (200 keV), where the ratio of calculated-to-measured values was 1.8. The reason for the discrepancies has not been determined. However, as stated above, the MCNP results actually applied relied on measured detector efficiencies. Thus, uncertainties in calculated detector efficiencies are not a factor in the analyses.

Results of the Shielding Analyses

Collimator Design - A number of collimator designs were evaluated. An open-faced arrangement with a variable amount of lead (Pb) shielding was determined to be best for this application. The modeling arrangement is shown in Figures 3 and 4. The dimensions of the face are 10.2 x 10.2 cm (5 x 5 in). With this opening, the detector is directly exposed to the whole cross section of the instrument trees over approximately a 30 cm (11.8 in) length. The collimators for two of the three detectors included some shielding to provide the capability of measuring a wide range of contamination levels. The shield thicknesses selected were 4.8 cm (1.9 in) and 7.6 cm (3 in); one detector had no shielding to provide maximum sensitivity. With the chosen detector shielding arrangement, there are approximately two decades of overlap between detectors (based on a useable detector range of 20 - 200,000 counts per second and a source

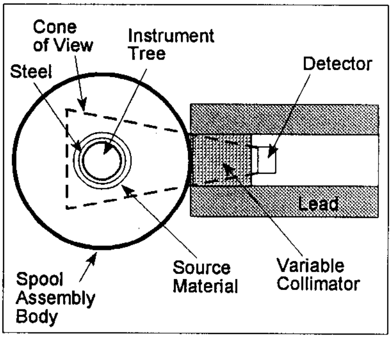

Figure 3 X-Y slice through the MCNP model of the FRRDS and Instrument Tree.

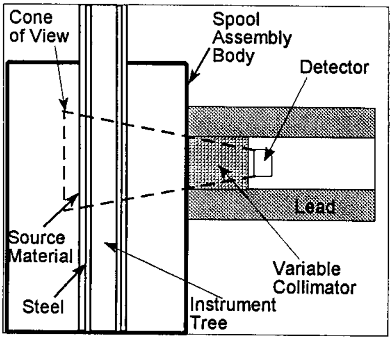

Figure 4 X-Z slice through the MCNP model of the FRRDS and Instrument Tree.

dominated by ^{137}Cs), as shown in Table 1.

Table 1. Computed Range of each Detector in Terms of ^{137}Cs Concentrations (Ci/ft) on Thermocouple Instrument Trees

Contamination Source	Lead Shielding Thickness in Collimator		
	0 cm	4.8 cm	7.6 cm
Liquid	6.2E-7 to 6.2E-3	1.4E-4 to 1.4E-0	2.6E-3 to 2.6E+1
Sludge	1.3E-7 to 1.3E-3	2.8E-5 to 2.8E-1	2.1E-4 to 2.1E-0

Adequacy of Shielding Provided by the Detector Housing - An analysis of the detector housing design concluded that it provides sufficient shielding under the assumption that the contamination remaining on the instrument tree after washing was not exceedingly high or nonuniform along the length of the IT. However, for the least-sensitive detector design, MCNP calculations indicated that a significant fraction of the detector signal could come from photons entering the detector chamber though the top if there was only the 5.1 cm (2 in) of top shielding provided by the housing, or from backscatter if the back side of the housing remained unshielded. This could have made interpretation of the count rate data difficult due to a high uncertainty in the radionuclide inventory inferred from the detector data, and could have limited the useful range of the detector.

The calculations showed that for the least-sensitive detector, the photon flux at the detector from backscatter could be as much as 26 (for the liquid source) to 45 (for the sludge source) times greater than the photon flux through the collimator, if the back of the detector housing remained unshielded. Consequently, the useful range of the detector would be reduced by back-scatter flux. The back-scatter flux was, however, at lower energy than the flux through the collimator, and the flux around the ^{137}Cs peak at 0.662 MeV was not affected. Because the back-scatter flux is dominated by lower-energy photons, 1 cm (.4 in) of lead shielding at the back of the detector housing effectively eliminated back-scatter flux.

If additional lead shielding was not added to the top of the least-sensitive detector housing, the detector count rate due to photons penetrating the top of the housing would exceed the count rate due to photons coming through the collimator. Unlike the backscatter photons, though, photons entering the detector chamber through the top of the housing would have an energy spectrum similar to that for photons entering though the collimator. This would have complicated the interpretation of measured count-rate data.

For the other two detectors, photons backscattering into the detector chamber, and photons entering the chamber through the top of the housing could also have been a problem if the axial distribution of the contamination after washing was very non-uniform. Without additional shielding, a relatively large, but localized deposit of radioactive material that was, for example, 60 cm (24 in.) above the elevation of a detector could produce a count rate in excess of the count rate produced by a thin film of contamination in front of the collimator. As a result of these analyses, lead shielding was added to the rear of the detector housing and to the top of the housing.

Correction Factors Applied to FRRDS Data - Detector calibration measurements were made in the field to determine the relationship between detector count rates and radionuclide inventory (Ci) as a function of photon energy. The geometry of the reference source used in these measurements was essentially a point source, which was not representative of a source expected to be distributed along the length of an IT. Consequently, MCNP calculations were made to determine factors to correct for differences in source geometries.

These calculations were done in pairs. The first model of each pair contained a mono-energetic point source with an arbitrary photon source rate (N photons/sec) simulating the calibration measurements. The second included a simulated IT with a mono-energetic source uniformly distributed as a thin annulus over the IT surface. The source strength in this model was set at N photons/sec per lineal foot. The ratio of computed detector count rates around the source energy (model 1 to model 2) then gave a factor to convert an FRRDS computer readout in curies to curies/foot of IT length. Each pair of calculations was repeated with a different source energy. The results of the MCNP calculations are shown in Table 2. Table 3 contains the correction factors derived from the data in Table 2 and applied to readings taken during the removal of the ITs.

Table 2. Normalized Detector Count Rate Data from MCNP Calculations of Models containing Point Sources and Distributed Sources

Photon Energy (KeV)	Model with Point Source			Model with Distributed Source		
	Collimator Shield Thickness			Collimator Shield Thickness		
	0.0 cm	4.8 cm	7.6 cm	0.0 cm	4.8 cm	7.6 cm
300	1.0(a)	(b)	(b)	0.612	(b)	(b)
400	1.0	(b)	(b)	0.608	(b)	(b)
583	1.0	1.40E-03	(b)	0.667	5.29E-04	(b)
662	1.0	3.62E-03	1.39E-04	0.675	1.87E-03	7.09E-05
861	1.0	1.54E-02	1.18E-03	0.709	8.93E-03	6.51E-04
1,621	1.0	6.54E-02	1.43E-02	0.738	4.46E-02	9.21E-03
2,615	1.0	1.05E-01	2.90E-02	0.926	8.33E-02	2.05E-02

(a) Count rate data at each energy were normalized to the value obtained for the detector with no shielding in the case with the point source.

(b) No data are given in these instances due to low count rates resulting in a large uncertainty in the MCNP computed data.

Data Processing for Waste Classification

Following the removal of an IT from the tank, the recorded data was processed using a spreadsheet to estimate the total ^{137}Cs activity on the tree. The isotope ^{137}Cs was used as the reference isotope because it is the only isotope consistently found in all layers of the tank waste. Table 4 shows a representative set of data from the intermediate range detector. The MCNP calculated correction factor and a calibration factor are applied to the raw data to calculate the corrected activity. Because the bottom sludge was assumed to be 46 cm (18 in) in the tank sludge, the total activity was separated into a total liquid phase activity and a total sludge phase activity.

Table 3. Factors to Correct FRRDS Readings (Ci) to Units of Ci/ft

Energy (KeV)	Collimator Shield Thickness		
	0.0 cm	4.8 cm	7.6 cm
300	1.63		
400	1.65		
583	1.50	2.65	
662	1.48	1.93	1.96
861	1.41	1.72	1.82
1,621	1.36	1.47	1.55
2,615	1.08	1.27	1.42

Table 4. Intermediate Range Detector Activity (Curies)

Crane position(ft)	Corrected Activity (Ci)	Crane position(ft)	Corrected Activity (Ci)
19.83	3.25E-04	37.24	3.64E-03
20.91	1.64E-03	38.39	2.89E-03
21.99	2.89E-03	38.78	3.21E-03
23.13	5.34E-03	40.88	2.17E-03
24.20	4.30E-03	42.10	2.54E-03
25.30	1.82E-03	43.22	1.57E-02
25.97	3.33E-03	44.19	9.84E-02
27.04	3.54E-03	44.68	5.72E-02
28.12	7.25E-03	45.88	5.36E-02
29.23	1.50E-02	47.04	3.66E-03
30.36	4.55E-02	48.23	1.13E-02
31.53	2.11E-02	49.44	1.52E-02
31.80	2.00E-02	51.65	2.70E-03
32.42	1.36E-02	52.85	2.22E-03
34.82	3.52E-03	53.96	2.22E-03
36.03	2.57E-03	56.59**	4.19E-03
		liquid activity	4.28E-01
		Sludge activity	4.19E-03
		Total activity	4.28E-01

* Corrected value is: (Measured Activity)*(MCNP correction factor)*(Calibration factor), ie. Activity*1.93*46, respectively.

** Portion of tree assumed to be immersed in sludge layer.

Concentrations of all other isotopes on the tree were estimated based on the analysis of tank waste samples and their calculated relationship to ^{137}Cs (Hetzer, 1995). The total ^{137}Cs activity used for this calculation was the total average corrected activity for the operable detectors. For waste disposal, the final data were reported in curies per cubic meter and nanocuries per gram.

Calibration and Performance

An acceptance test was performed on the FRRDS detectors. Several measurements were made on each detector to assure compliance with efficiency, resolution and throughput. Two primary traceable radioactive sources of ^{137}Cs and ^{228}Th were used. The cesium source was used for resolution and efficiency tests. Both sources were used in determining throughput and high count rate effects. The thorium source was placed at a stationary position and its 2614 keV photon was monitored. The cesium source was progressively moved closer to the detectors until count rate was sufficient to overload the system. Figures 5, 6, and 7 show stability of resolution, peak shift and activity recovery respectively as the input count rate was increased. The primary observation is that the system shows changes but software correction of the data based on the pulser information provides accurate results.

Subsequent to acceptance, the detectors were placed into the FRRDS assembly for

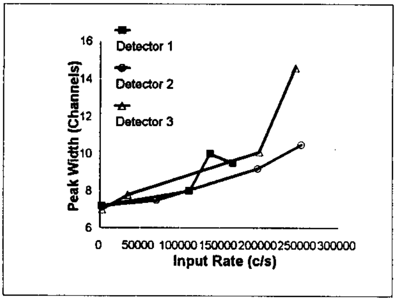

Figure 5. Effect of input count rate on observed resolution at 2614 keV.

empirical calibration. A traceable thorium source was placed in the center of the spool piece in line with the detectors. Data were acquired sufficient to generate an energy versus efficiency curve for each detector over the energy range encompassing 200 to 2614 keV. These data were then adjusted from point to line geometry and additional expected overhead 'shine' using the MCNP code as discussed above. Figure 8 shows the final efficiency curves used to report field measured nuclide quantities. In this figure, detector 3 is attenuated nearly five orders of magnitude for the most abundant nuclide (^{137}Cs) energies by the collimator shielding.

During operation, the system was set to acquire data on a one minute real time basis. At the start, only the least shielded detector showed response for this short period. As contamination increased, the second and third detectors responded with the first becoming over saturated as the secondaries started up. This response and the correlation of the three detectors is shown in Figure 9 for ^{137}CS.

It was expected that under severe conditions false positives for illogical nuclides would be reported due to random summing events. Therefore, a filter program was prepared which accepted the primary data reduction output, merged crane position data, and generated a list

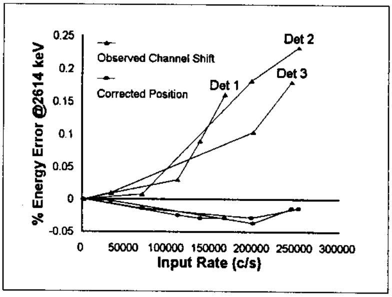

Figure 6 Energy shift of 2614 keV gamma photopeak with increasing low energy count rate, observed and pulser corrected.

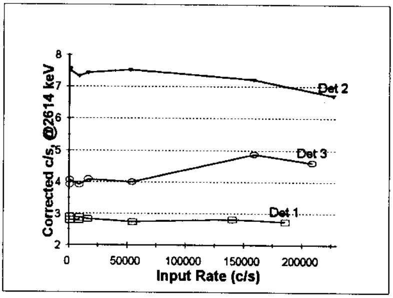

Figure 7 Pulser corrected count rate stability.

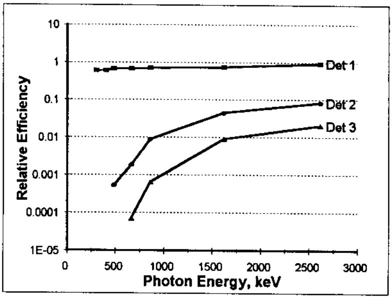

Figure 8 Detector efficiency curves corrected for line source distribution and external gamma shine.

suitable for final MCNP correction operations. It was found that the only logical, consistent, and significant nuclides were ^{60}Co, ^{137}CS, and ^{154}Eu. Other expected long-lived low energy nuclides such as ^{241}Am are hidden by the intense abundance of ^{137}Cs. An example profile of these nuclides is shown in Figure 10.

As a final aspect of the system performance, Figure 11 shows the affect of the high pressure washer on the contamination. Dose rate meters were placed on the system, one at the top of the tank riser (see Figure 2) and the second above the wash system. The system was able to reduce radioactive contamination by twenty fold.

Summary

The FRRDS system demonstrates the ability to apply laboratory quality gamma spectroscopy in extreme field operations. The design anticipated a need for large dynamic range of radiation exposure as shown in the actual usage. Theoretical modeling, primary calibration, and actual operations show a consistency in the design and performance. The demonstrated performance of the system is only possible through the combined pulser injection and software correction system.

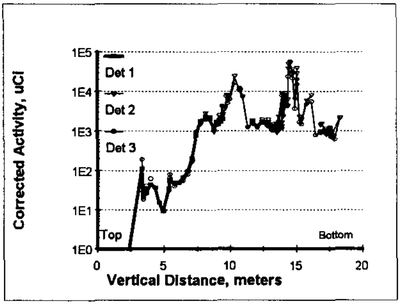

Figure 9. Correlation of detector response along thermocouple tree during removal. Detector 1 is inoperable at 9.1-12 m (30-38 ft) and 14-16 m (45-54 ft) intervals due to radiation intensity.

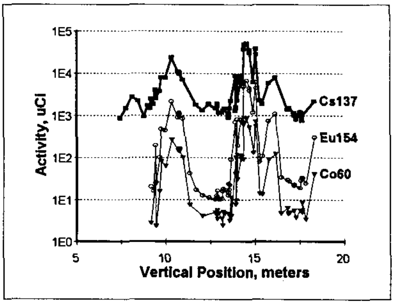

Figure 10 Example radiation profile for primary detectable radionuclides for a thermocouple tree removal.

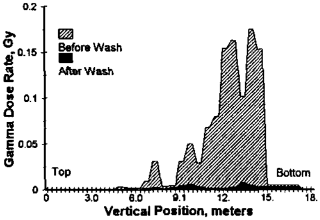

Figure 11 Comparison of radiation dose rates before and after high pressure wash of a thermocouple tree.

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