Use of Field High Resolution Gamma Spectroscopy during Decontamination and Decommissioning of Nuclear Facilities - 9220

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ABSTRACT

This paper describes the utilization and evolution of field high-resolution gamma spectroscopy (field-HRGS) during decontamination and decommissioning (D&D) of a nuclear facility; beginning with characterization of drummed and boxed radioactive waste for purpose of shipment and disposal, and ending with survey and analysis of radioactive material held-up in building fixtures and structures leading to improvements in radiation safety, environmental protection, waste disposal, and regulatory compliance. This technology also provides detailed documentation of regulatory compliant clean-up and disposal activities.

Compared to other radiation detection technologies, HRGS provides the best gamma radionuclide identification capabilities for both naturally occurring radionuclides such as U-238, U-235, U-234, Th-232, Th-228, Ra-226 and their daughter products as well as other radionuclides (Pu-239, Am-241, Ba-133, Cs-137, Co-60, etc.) associated with human activities. No other technique can match the ability to quantify gamma radionuclides on an activity, mass, and concentration basis in the field as well as HRGS.

HRGS is a powerful analytical tool often overlooked by industry in favor of other less costly and less complicated measurement techniques such as sodium iodide (NaI) or cadmium zinc telluride (CZT) survey.

When used properly - in a coordinated effort during D&D operations, field-HRGS survey and analysis can provide large benefits in radiation and environmental protection, waste reduction, waste disposal, and regulatory compliance with anticipated lowering of risk and overall savings in cost.

INTRODUCTION

Decontamination and decommissioning of nuclear facilities is a risky, costly and time consuming process. Being able to accurately determine the type, extent, and quantity of radiological contamination from beginning to end has many benefits. Field-HRGS proved to be a powerful analytical tool for this purpose, as presented in this paper.

Lessons learned from characterization of transuranic (TRU) and low-level radioactive waste (LLRW) containers for shipment and disposal were applied to expand the use of this technology to other areas of the D&D process.

EXPERIENCE

Much of the experience gained and lessons learned as presented in this paper are the result of D&D activities conducted at the former DOE Rocky Flats plutonium processing facility located near Golden, Colorado. From 2003 through 2005, the author performed over 1500 documented "in-field" HRGS surveys and analysis on a wide variety of radioactive contaminated materials to include interior fixtures and areas within plutonium process Buildings 771, 371, and 559.

A sampling of the types of materials and objects analyzed include:

Waste Containers
55-gallon waste drums (TRU, LLRW)
SWB boxes (TRU)
Intermodal waste containers (LLRW)

Building Fixtures
Glove boxes
Criticality drains
Overhead process pipes and lines
Waste and exhaust pipes, stacks, and filter plenums
Waste tanks (above and below ground)
Remote handlers

Materials
Concrete floors, walls, and ceilings
HEPA filters
Ion exchange resins and fluoride wastes
Metals, glass, and PVC
Soils

Survey and Analysis Equipment

Survey equipment used during this period consisted of portable liquid-nitrogen-cooled high purity, high resolution germanium (HPGe) detectors N-Type (25% to 80% relative efficiency), DigiDart^{tm} (HV power supply, MCA, and supporting electronics), Windows based laptop computers, detector orientation-adjustable wheeled survey carts, lead shielding with collimation, hand held sodium iodide (NaI) survey meters, spectrum processing and analysis software such as Maestro^{tm}, Isotopics^{tm}, ISOCS^{tm}, and SNAP^{tm}.

Use of this equipment provided benefits in the following areas:

- Enhanced radionuclide identification and waste classification capabilities
- Enhanced quantification (activity, mass, concentration) of radioactive material
- Contamination depth profiling in concrete
- Safer airborne and high contamination area survey and analysis
- TRU waste reduction
- Verification of clean-up activities
- Detailed supporting documentation

Radionuclides of Interest

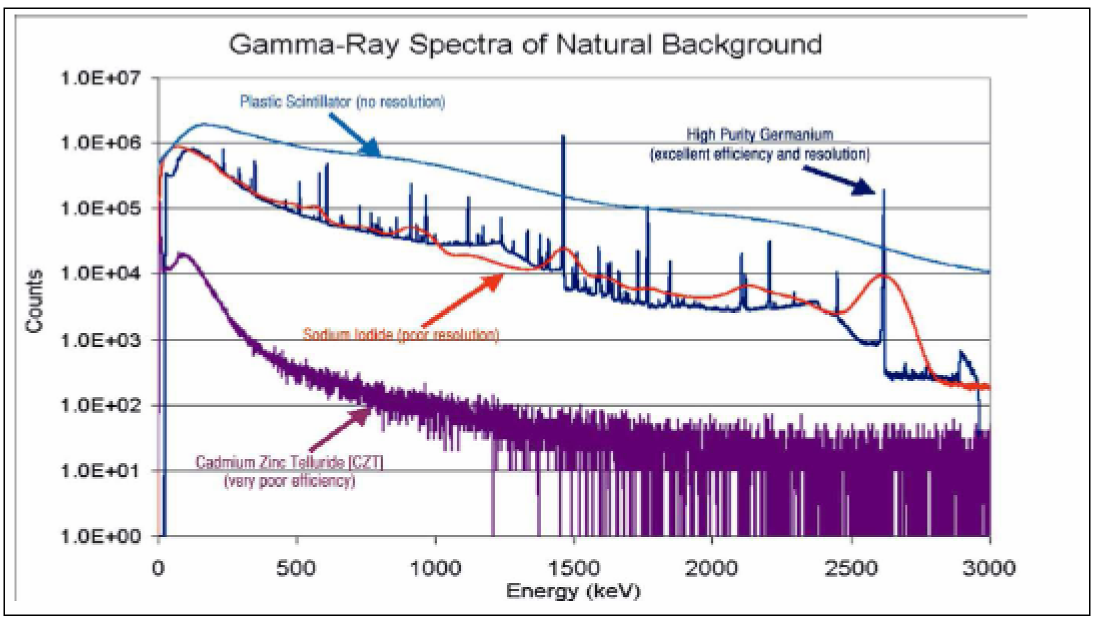
Monitoring of specific gamma-emitting radionuclides during this period included: Am-241, Ba-133, Co-60, Cs-137, Cs-134, Np-237, Pu-239, Ra-226, Th-234, Th-228, U-238, U-235, and U-234.

FIELD HIGH RESOLUTION GAMMA SPECTROSCOPY

What is field high resolution gamma spectroscopy (field-HRGS)?

For purpose of this presentation, field-HRGS is defined by existing technology that provides the ability to collect and analyze high-resolution gamma spectroscopy (HRGS) spectra in the field for identification and quantification of radioactive materials on an individual radionuclide basis for a variety of materials under a variety of survey conditions. This means the ability to collect detailed high-resolution gamma spectra, identify individual radionuclides of interest, detect normal background radionuclides, and account for survey parameters (geometry and attenuation) of source material to achieve reasonable estimates of radionuclide quantities and concentrations. The equipment allows for adjustment of count-time, distance, detector orientation, shielding, and collimation on the spot to achieve maximum efficiency and reliable analytical results. In addition for purpose of this presentation, HRGS is defined as a process that provides gamma photo-peak resolution that is equivalent to the resolution associated with high-purity germanium (HPGe) type detector systems, which can achieve photo-peak resolution of less than 3 kilo-electron volt (kev) at full-width half-maximum (FWHM) for Cs-137 at 662 kev.

Figure 1 provides an illustration of a comparison of the detection efficiency and photo-peak resolution obtained from a HRGS High Purity Germanium detector system verses other radiation detection systems.



Fiqure 1 – Published by Ortec. Obtained from white paper titled “Why High-Purity Germanium Radiation Detection Technology is Superior to Other Detector Technologies for Isotope Identification”.

Field-HRGS Detection Limits

Field-HRGS detection limits are dependent upon detector efficiency, source material matrix and survey parameters such as geometry and attenuation. When used appropriately, field-HRGS can provide reliable detection limits that are quantifiable and appropriate for the materials surveyed. For example, the detection limit for Cs-137 contamination within a 1000 cubic centimeter volume (10x10x10 cm) of soil at a distance of 15 cm is approximately 30 bequerels (900 pCi or 0.6 pCi/g) for a 60 minute count time and detector efficiency over 50%. Gamma spectrum analysis software should be able to determine detection limits under a variety of field survey conditions.

TECHNOLOGY EVOLVED

One of the more important lessons learned during D&D activities at Rocky Flats was the flexibility of field-HRGS with realization of its full potential in identifying and quantifying radioactive contamination from high levels of TRU to normal background radiation levels within a wide variety of material matrices under a myriad of conditions, from the early stages of D&D through final verification of meeting regulatory release criteria.

The use of this technology evolved throughout the D&D process providing benefits related to radiation and environmental protection, waste reduction, and regulatory compliance resulting in more efficient D&D operations.

Waste Containers – Regulatory Compliance

Initially, field-HRGS survey and analysis was relegated to identification and quantification of transuranic (TRU) and low-level radioactive waste (LLRW) contained in standard 55-gallon drums and SWB waste boxes for purpose of regulatory compliant shipping and disposal.

Fiqure 2 – Field-HRGS survey and analysis of SWB TRU container in Building 371 prior to shipment to WIPP.

Dismantled Equipment – Waste Reduction

As D&D progressed, field-HRGS survey and analysis migrated to cut sections of contaminated equipment and material after removal, and prior to or during placement in disposal containers. This proved to be beneficial providing more accurate analysis with reduction in the need to analyze filled drums and SWB boxes, which are more difficult to characterize. Further benefit was realized under conditions where field-HRGS survey and analysis was able to distinguish areas of transuranic (TRU) verses low-level radioactive contamination for further material-waste reduction prior to containerization.

High Contamination and Airborne Radioactivity Areas – Radiation Protection

Field-HRGS was especially useful in providing analytical results for radioactive materials within high-level radioactive waste reduction enclosures, glove boxes, waste tanks, and HEPA air-filter plenums.

Figure 3 – Field-HRGS survey and analysis of TRU waste SWB container while still attached to an airborne HCA material reduction area and glovebox.

Measurements were performed on the outside of these structures at safe distance and away from airborne contamination, reducing the risk of exposure to the operator and contamination of survey equipment. Detector-to-source distances were easily adjusted to provide additional radiation protection for survey personnel. Survey and analysis provided reliable radionuclide quantification for determination of TRU radioactive waste classification with assurance of maintaining criticality limits.

Building Structures – Waste Reduction, Radiation and Environmental Protection

As interior fixtures and structures were removed from a facility, exposed concrete walls, floors and ceilings were surveyed to locate residual areas of contamination and to determine depth of contamination. This information was used to aid in removal of contaminated paint and concrete to levels below regulatory concern thus reducing volume of radioactive waste. In addition, follow-up measurements were performed after decontamination to document achievement of radiation protection and release standards.

Special Surveys and Analysis – Waste Reduction, Environmental and Radiation Protection

The so called “Infinity Room” in Building 771 is a good example of how field-HRGS can be put to use in identifying and quantifying radioactive contamination under the most adverse conditions. This room was designated as an airborne HCA area for many years with high levels of radioactive contamination distributed throughout the room, on the floor, walls, ceiling, and two concrete pedestals.

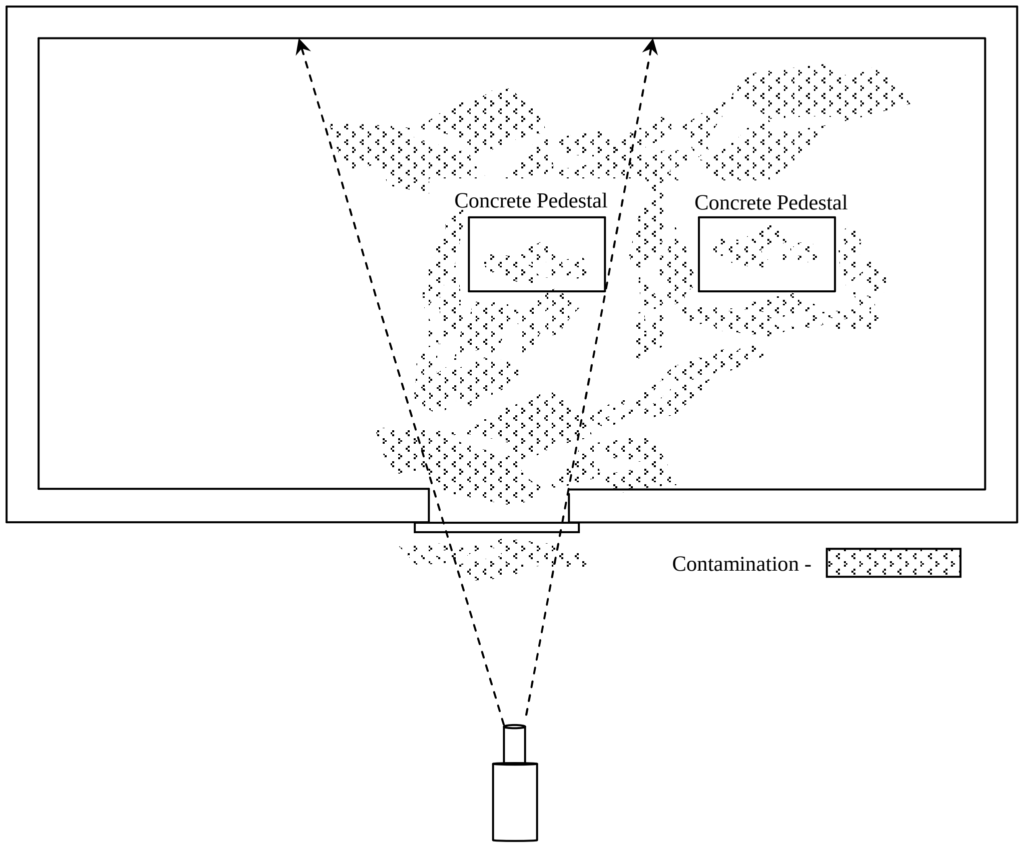


Figure 4 – Schematic (not to scale) of Building 771 Pump Room (Infinity Room – Airborne High Contamination Area).

Prior to field-HRGS survey and analysis, not much was known about the extent and quantity of contamination, other than process knowledge which indicated the presence of plutonium and americium, while radiological surveys indicated an airborne HCA.

During the D&D process, preliminary field-HRGS survey and analysis was conducted from a safe distance, outside of the room looking through the doorway which was sealed shut with a 0.5 cm thick metal plate, as depicted in Figure 4.

Modeling parameters (geometry, attenuation, etc.) for quantification of radioactive contamination were estimated based on scaled floor-plan schematics, since visual observation and measurement of the interior was not feasible. Gamma spectral analysis verified the presence of Pu-239 and Am-241 contaminates with an estimate of at least 12 grams of Pu-239 and several grams Am-241 contamination within the detector field of view.

Additional information was needed to determine the best approach for D&D of this area, so follow-up field-HRGS survey and analysis of the floor, pedestals, walls, and ceiling was performed inside the room after mitigation of airborne contamination. Spectral analysis identified areas of TRU quantities of plutonium and americium contaminated concrete material on and around the pedestals including many parts of the floor to depths approaching 6 inches. Wall areas were able to be classified as both TRU and LLRW while many areas of the ceiling were determined to be below regulatory concern. Final estimates indicated at least 20 grams of Pu-239 contamination within the room with a large volume of TRU (>100nCi/g) contaminated concrete waste. Third-party analysis of concrete samples pulled from the room, verified field-HRGS estimated quantities and concentrations for plutonium and americium. Based upon these results of analysis, a D&D plan was developed accordingly.

Putting It Altogether – Coordinated Survey and Analysis

In the fall of 2004, lessons learned from D&D of Buildings 771/774 and 371/374 were applied to Building 559 (high-level analytical laboratory) in a more coordinated effort. Unlike previous operations, Building 559 field-HRGS survey and analysis began with existing structures and fixed equipment still in place (prior to removal) which provided valuable information on the location and extent of radioactive contamination prior to D&D removal activities.

This coordinated effort proved to be most beneficial. Project managers were able to pre-determine volumes of TRU, LLRW, and below regulatory concern (BRC) material prior to commencement of D&D activities with gains in safety, environmental protection, regulatory compliance, and reduction of volume of TRU and LLRW waste. In turn, operations personnel had access to detailed documentation of the type, extent, and quantity of radioactive contamination prior to material disturbance, and could do a better job of coordinating D&D activities. After fixtures were removed, exposed building walls, floors, and ceilings were surveyed to locate areas of residual contamination requiring further mitigation.

This coordinated effort reduced the number of surveys needed to be performed on filled waste containers and dismantled sections of interior fixtures such as overhead ducts and pipes. In the end, final "close-out" surveys were performed using the same equipment to verify and document successful clean-up activities.

Added Benefits - Detailed Documentation

All field-HRGS survey and analysis data was documented and archived in electronic format, including full-detailed high-resolution gamma spectra which allowed for complete data review, verification, and validation. This proved to be helpful when initial results were questioned or when additional information became available after a survey was completed, providing an opportunity for re-verification or re-analysis of materials that were no longer available for re-survey.

Final benefits were realized as regulators and stakeholders were presented with detailed radionuclide identification and quantification documentation that demonstrated successful and regulatory compliant clean-up activities. High resolution gamma spectra showing radionuclide contamination and background radiation before, during, and after D&D can provide strong "cradle to grave" evidence of success.

Detailed documentation includes:

- Instrument calibrations
- Instrument daily quality control checks
- Date/ time and location of surveys
- Description of objects surveyed
- Survey modeling data (geometry, attenuation, etc.)

- Detailed gamma spectra
- Raw data and calculations
- Final report of analysis with determination of estimated error

Cost Benefits

Overall benefits and cost savings gained in risk management, radiation and environmental protection, waste reduction, and regulatory compliance were hard to quantify. Although, if we consider the total estimated costs of D&D for Buildings 771, 371, and 559, the labor and materials expense associated with field-HRGS survey and analysis amounted to no more than $1.5 million which is a fraction (less than 1%) of the total estimated "Facility Decommissioning Cost Model" cost of $168 million for these three facilities. Unfortunately, no cost-savings estimates were determined from "coordinated survey and analysis" efforts associated with any of the D&D operations.

CONCLUSION

Field-HRGS survey and analysis proved to be an effective tool for characterization of contaminated material associated with nuclear material operations. Coordinated survey and analysis of material "in-place" (prior to disturbance and removal) during D&D operations can provide gains in radiation and environmental protection, waste reduction, and regulatory compliance resulting in more planned and less reactionary operations. Radioactive materials can be surveyed at safe distance without the need for special radiation protection measures. The technology is flexible enough to provide acceptable analytical results of analysis on a wide variety of materials under a wide variety of field conditions from high to normal background levels of radioactivity.

In closing, field-HRGS is a powerful and reliable analytical tool that can provide detailed documentation of identity and quantity of nuclear material on an activity (Bq), mass (gram), and concentration (Bq/g) basis, if used properly and to its full potential.

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