PRESHIPMENT ON SITE CHARACTERIZATION OF HISTORICAL LLRW AT BROOKHAVEN NATIONAL LABORATORY

K. A. Carney, CHMM, Brookhaven National Laboratory
J. Burn, PhD, Safety & Ecology Corporation
S. Jones, Safety & Ecology Corporation
M. Kaye, Safety & Ecology Corporation
F. Myers, Safety & Ecology Corporation

ABSTRACT

Waste containers at Brookhaven National Laboratory (BNL) were characterized onsite using an In-Situ Object Counting System (ISOCS) for shipment to a processing facility for ultimate disposal. The waste had been collected from BNL operations over several years with a wide variety of radionuclide contaminants. The exact origin of the contents of the containers could not be verified with the certainty necessary to characterize the material for shipment. In addition, the waste did not necessarily originate from a single source and was not homogenously mixed in the containers.

In order to ship the material, the radionuclides in each container had to be identified and quantified in order to meet the waste acceptance criteria for the recipient. The separate packaging of different waste items within the containers and their diverse process origins made obtaining an intrusive representative sample virtually impossible. Recent advances in in-situ field gamma spectroscopy led to the determination that the containers could be accurately measured individually to characterize their contents. These measurements along with historical scaling for low energy or non-gamma emitting isotopes allowed the shipping broker to accurately manifest the waste for shipment.

The containers varied in physical construction and included metal boxes, concrete vaults, shielded transport "pigs", and Sea-Lands. The physical form of the material was determined by historical records of each container and/or by direct inspection when the data was not available or was in doubt. These records and inspections were also used to confirm that the material was properly characterized in regards to other hazardous constituents. The determination of the physical form was also critical in the modeling of the containers using the ISOCS software. Random intrusive samples were also taken from a percentage of the containers and laboratory analyzed to backup the characterization activities.

The containers were analyzed using a Canberra REGe detector with a 70% efficiency. This allowed for reduced count times and accurate measurement of gamma energies down to around 10 keV. The containers typically had significant quantities of radionuclides. This along with the high efficiency of the detector generally resulted in count times of 15 minutes or less per spectra. The number of spectra collected range from 2-6 depending on the geometry of the container. This resulted in a process rate of 6-8 containers in a typical ten-hour workday. Spectra analysis,

scaling, and modeling then were performed on each container to determine the radionuclide concentrations.

INTRODUCTION

The waste characterization and removal is one of the activities ongoing for the Bechtel-managed Environmental Restoration Division at BNL. The project is part of the overall cleanup of the old waste operations facility located in and around Bldg. 445, commonly referred to as the "boneyard".

Originally an ammunition storage area when BNL was the U.S. Army's Fort Upton earlier last century, the boneyard was converted to the operational waste management facility for BNL several decades ago. The boneyard was replaced by a new state of the art facility that was constructed and placed in operation in the mid 1990's.

The first stage of the cleanup at the bone yard involved the characterization, packaging, and shipment of a huge amount of waste from the facility. This included more that 225 B-25 and B-12 containers, 30+ concrete vaults ranging from 6-17" thick, several Sea-Land containers, and more than 100 sources no longer used at BNL. The waste varied in form and included typical dry active waste (DAW), construction debris, soil, and even fuel rod end pieces. The activity in the containers varied over a wide range, from DAW with activities at the lower range of detection to fuel ends with several Curies per container. The sources varied in activity from the millicurie range to a strontium-90 source with over 20,000 Curies and cobalt-60 and cesium-137 sources of over 5000 Curies each.

The project included waste characterization, shipment, processing, and disposal along with radiological control and health and safety functions. This included the use of state of the art methodologies for the measurement and characterization of the waste packages and items. The project team employed the ISOCS system, an in-situ gamma spectroscopy system, to analyze the packages and give the type and quantity of radionuclides present in the waste. This was done without the need to perform intrusive operations on the packages, thereby minimizing hazards to personnel and allowing for significantly more efficiency. The project, originally scheduled for completion in mid-2001, has increased in scope by some 30%, yet will still be completed more than two months ahead of schedule.

WASTE FORM DESCRIPTION

In order to model the containers accurately, the physical characteristics of the container and the waste had to be determined. The containers were measured in the field and drawings were obtained from BNL and the container manufacturers to verify construction material and dimensions of each container's sidewalls.

The historical information was reviewed for each container and it's physical content determined. Direct inspection was performed on container contents for which there was no historical information or when the data was in doubt. The material in the containers varied from typical DAW items such as paper, protective clothing, laboratory waste, construction debris (wood, concrete, etc.), to system components such as piping and valves. One stream processed was aluminum fuel rod ends that had been removed from the active fuel area of one of the research reactors at BNL and stored in concrete shield vaults due to their high activity. The volume and the makeup of the material along with the container characteristics were necessary for the modeling using the ISOCS system.

INSTRUMENTATION

The principal instrumentation used for the project included Canberra ISOCS and LabSOCS units and a Tennelec low background counting system. The ISOCS was used for the field measurement, while the LabSOCS and Tennelec were used primarily for analysis of samples involved in the radiological control portions of the project.

The ISOCS system consisted of the following major components: a 70% efficient REGe "ISOCS Characterized" Germanium Detector, a set of shields and collimators on a portable cart, and a portable InSpector 2000 acquisition electronic unit. The detector for the system is characterized by the manufacturer using the well-known MCNP Monte Carlo modeling code. Specifically, the radiation response profile of the detector to be used is determined for a 50 meter sphere about the detector over a 50 keV through 7 MeV energy range. The results of this characterization are then used by the ISOCS software to provide accurate quantitative data without the need to acquire a variety of calibration sources plus hours of calibration time and effort. ISOCS and traditional efficiency calibrations typically agree within a few percent. The LabSOCS system is essentially an ISOCS unit configured in a typical fixed laboratory setting. This unit can be used with traditional sources/geometries or with the modeling techniques the ISOCS system offers. The use of both systems allow the simultaneous acquisition and modeling of spectra that greatly increased the production during the project. (Generally, the modeling and data analysis effort requires more time than the actual sample spectra acquisition.) Both of the spectroscopy systems were operated using IBM-compatible PCs running Canberra's PROcount and associated Genie-2000 software in Windows 98.

While the typical ISOCS detector is a coaxial Germanium with a relative efficiency of 40-60%, a REGe (reverse electrode Germanium) detector was selected for this project. The REGe is similar in geometry to other coaxial germanium detectors with one important difference. The electrodes of the REGe are opposite from the conventional coaxial detector in that the p-type electrode, (ion-implanted boron) is on the outside, and the n-type contact (diffused lithium) is on the inside. There are two advantages to this electrode arrangement; window thickness and radiation damage resistance. A thin, rugged carbon composite window was used due to the field activities. The detector arrangement resulted in a range of approximately 10 keV to 10 MeV. Experimental evidence also suggests that the REGe detector may be 10 times as resistant to damage as conventional Coaxial Ge detectors.

The ISOCS system was also equipped with a shield package included both 2.5 cm (1 inch) and 5 cm (2 inch) lead shield assemblies with collimators of 30°, 90°, and 180° to minimize interfering radiation and limit the field of view.

WASTE SPECTRA ACQUISITION, MODELING AND ANALYSIS

The containers were located in a fenced in area with background radiation levels ranging from 10 microrem to several millirem per hour. The area was extensively surveyed to allow for the spectra acquisition to be performed in the lowest possible background. The location for the majority of the spectra acquisition was approximately 10 microrem/hr. The collimated arrangement of the detector also required that detector orientation be considered. To further decrease background radioactivity, side collimation was always used. While, the determination of the number and orientation of the spectra will be further discussed in the modeling section below, the basic sequence of daily operations was to perform and verify background and check standard quality control measurements and then sample measurements. The sample backgrounds were performed in the same detector to sample configuration including detector orientation to other potential sources at the facility and shield configurations. The wide range of the sample activities required that several typical geometry-and-collimation configurations be developed for each package type.

This was done by performing geometrical calculations to ensure the field of view of the detector was sufficient in each of the shield/package locations (i.e. distance to the detector). The operator would then choose the best configuration for each package with the minimal use of front shielding on the detector. This was based on limiting detector dead time $\leq$10%. A typical laboratory standard detection limit maximum of 1 pCi/g for cesium-137 was used to determine count time. Initial counts and spectra analysis indicated that a count time of 15 minutes per spectra was sufficient to achieve this level of sensitivity. The high typical waste activity and the high efficiency of the detector also affected this determination. Figure 1 below shows the collection of a typical spectrum.

ISOCS modeling of each container was performed independently of data collection. Empirical data describing each container was collected from a combination of historical records, when available, and physical inspection. The location of the waste container relative to the detector, the materials of construction and wall thickness of the waste container, and the net mass, volume, density, and approximate elemental composition of the contents were gathered for use in the ISOCS modeling process.

Fig. 1. Spectrum Acquisition on B-25 Waste Container

The spectra were then analyzed using the PROcount-2000 software for isotopic identification and quantitation. This data, along with historical scaling previously discussed, was then used for shipping purposes. The isotopic identification was one of the most difficult portions of the entire project. The research activities at BNL have resulted in the production of radionuclides that are extremely diverse. They included activation products from reactor and accelerator operations, naturally occurring isotopes, reactor fuel, fission products, and medical isotopes. The library for the analysis was developed from broad library templates supplied with the software and then customized based on sample spectra and gamma production tables. The high activity of the waste also resulted in the production of significant production of characteristic x-rays that further complicated this process.

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