Validation of Non-Invasive Waste Assay System (Gamma Box Counter) Performance at AECL Whiteshell Laboratories – 13136

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ABSTRACT

Low-level radioactive waste (LLW) in solid form, resulting from decommissioning and operations activities at AECL's Whiteshell Laboratories (WL), is packaged in B-25 and B-1000 standard waste containers and characterized before it is shipped to an on-site interim storage facility, pending AECL decisions on long term management of its LLW. Assay of the waste packages before shipment contributes to an inventory of the interim storage facility and provides data to support acceptance at a future repository. A key characterization step is a gamma spectrometric measurement carried out under standard conditions using an automated, multi-detector Waste Assay System (WAS), purchased from Antech Corporation. A combination of ORTEC gamma acquisition software and custom software is used in this system to incorporate multiple measurements from two collimated high-resolution detectors. The software corrects the intensities of the gamma spectral lines for geometry and attenuation, and generates a table of calculated activities or limits of detection for a user-defined list of radioisotopes that may potentially be present.

Validation of WAS performance was a prerequisite to routine operation. Documentation of the validation process provides assurance of the quality of the results produced, which may be needed one or two decades after they were generated. Aspects of the validation included setting up a quality control routine, measurements of standard point sources in reproducible positions, study of the gamma background, optimization of user-selectable software parameters, investigation of the effect of non-uniform distribution of materials and radionuclides, and comparison of results with measurements made using other gamma detector systems designed to assay bulk materials.

The following key components of the validation process have been established. A daily quality control routine has been instituted, to verify stability of the gamma detector operation and the background levels. A test box containing plywood (specific gravity 0.5) with holes for reproducible positioning of known point sources has been constructed and used in several configurations. Gamma spectra have been processed multiple times with user-adjustable parameters set to a variety of values to gain agreement on the most appropriate settings. A similar optimization process was followed with the custom software provided by the instrument manufacturer to reach agreement on the most stable settings for reliable results. Measurements made with the Canberra In-Situ Object Counting System (ISOCS) instrument were compared with the WAS results as part of an estimate of instrument precision and accuracy.

The validation report will conclude with recommendations for the configuration, appropriate use and limitations of the Waste Assay System.

INTRODUCTION

WL Decommissioning Strategy

Since 1963, AECL has owned and operated Whiteshell Laboratories (WL), a nuclear research establishment located in the Canadian province of Manitoba about 100 km northeast of Winnipeg. The site is comprised of a number of nuclear and non-nuclear facilities, including a 60-MW test reactor (WR-1, operated 1965-1985), the Shielded Facilities (SF), various research laboratories, and liquid and solid radioactive waste management facilities. In the late 1990s, a decision was made to end the research activities and begin decommissioning the facilities, in some cases placing them into safe storage with surveillance. In 2006, the Governmental of Canada adopted a new long-term strategy to deal with the country's nuclear legacy liabilities over a 70-year period. The Nuclear Legacy Liabilities Program (NLLP) gave AECL the mandate to proceed with an accelerated schedule to fully decommission the WL site with the expectation to reduce public risk and operational costs as soon as reasonably achievable, and to align decommissioning work with international best practices [1].

Need for Waste Assays

Low-level radioactive waste in solid form resulting from decommissioning and operations activities at AECL's WL is packaged in B-25 and B-1000 standard containers. An on-site interim storage facility called SMAGS (Shielded Modular Above-Ground Storage) has been constructed to hold the waste, pending decisions on long term management of LLW. Assay of the waste container before shipment to SMAGS provides a method of verifying that the waste acceptance criteria for the facility have been met, identifies the container's contribution to the total inventory of that interim storage facility, and provides data to support acceptance of the waste at a future repository.

Assay Methodology

A key characterization step is a gamma spectrometric measurement to confirm the radiological characteristics of the waste contents. The SMAGS acceptance criteria include an upper limit on container dose rate as well as an upper limit on fissile material content. Measurement of gamma-emitting radionuclides can be used together with information on composition of site waste streams to gain a reasonably comprehensive understanding of the waste characteristics. In particular, fission product and actinide yields, as well as historical information on laboratory activities, have been folded into a reliable description of expected radiological content of the waste. For non-gamma-emitting nuclides, the approach has been to use scaling factors established through analytical investigations of both routine and non-routine waste streams.

INSTRUMENTATION AND SOFTWARE

The Waste Assay System (WAS) at WL has been designed [2] to produce an estimate of the radionuclide inventory of waste containers destined for storage in the WL Waste Management Area. The containers hold decommissioning or operational solid waste known to be contaminated with radioactive material. The WAS calculates the radionuclide inventory based on spectrometric measurements of the gamma field outside the container. Steel boxes of standard size (approximately 1.2 by 1.2 by 1.8 m) are used to contain waste packages. These are known as B-25 or B-1000 containers, depending on details of their manufacture (see Figure 1).

Figure 1. B-1000 container and Waste Assay System (Box Counter)

The WAS (also known colloquially as the Box Counter) was purchased from Antech, Inc. as a turn-key instrument including both custom hardware and software. To assay a waste container, an operator places it on the instrument platform and enters relevant details of its nature and contents into the operating software. The instrument then positions both the platform and the two gamma detectors in several pre-specified arrangements to acquire up to twelve gamma spectra, two at a time. Based on detector dimensions and user-performed calibration information, the WAS calculates the radionuclide content of the container and generates a report. A key assumption of the calculations is that both the radionuclides and the bulk material ("matrix") in the container are more or less uniformly distributed within the detector field of view at each position. Non-uniform distribution of materials and radionuclides, especially the presence of "hot spots" within the container, has a significant effect on the calculated total and mean activity of the container.

Hardware Description (detectors, electronics, platform)

In Figure 1, the box is shown in position on the platform, which includes both a horizontal transport mechanism and a load cell under the platform. It is designed to move boxes weighing up to 6 Mg horizontally to one or more positions so that gamma spectroscopy measurements can be performed. In the photograph the two detector platforms can be seen mounted on two lifting pillars. Each platform holds one germanium detector, with its acquisition electronics, collimator, shielding, and electromechanical cooling. The vertical position of each detector can be changed under computer control. By moving both the box and the detectors any measurement position on each side of the box can be accessed. Typically a B-25 box is measured using 6 detector positions on each side, 3 positions above and 3 below the mid plane of the box, and 30-minute spectrum acquisitions per position.

Software Description

The top-level software, called BoxCounter, controls box and detector movements through programmable logic controllers (PLCs), as well as coordinating the operation of the data-acquisition and data-analysis modules. Gamma ray spectra are acquired and analyzed using the ORTEC software GammaVision-32 [3] for each box measurement position using the ENV32 analysis module. Following the spectrum analysis, ANTECH IsoCorr software calculates the attenuation correction for each volume element in the volume of the box seen by the detector for each separate box measurement position. For each box measurement, IsoCorr uses the density derived from the load cell reading and the container dimensions, together with the detector response from the GammaVision analysis, to calculate the total activity assuming uniform attenuation over the volume. Separate box measurement results are averaged to get total activity for the entire box. The final report is printed, and the entire workspace, including spectra, calibrations, and intermediate results, is saved as a zip file.

Instrument Calibration

The detector efficiency calibration is performed using an Eu-152 certified source placed at a fixed distance from the centreline of each detector face. The calibration is performed using the GammaVision-32 software and the resulting files are used by IsoCorr as the basis for correcting the acquired assay spectra for the container geometry and matrix. Energy calibrations are also performed using the Eu-152 calibration source either manually using GammaVision-32 or automatically from within the BoxCounter software.

Quality Control Program

Prior to each use, a check of the energy and efficiency calibration is performed for both detectors simultaneously, using a Eu-152 source placed in a reproducible position. Automatically generated control charts indicate if any parameter is outside specifications. Two types of background checks are also performed: a 5-minute measurement daily to detect obvious contamination, and a 16-hour measurement periodically to determine the appropriate data to subtract from box spectra. See below for more details on background measurements.

VALIDATION PARAMETERS

This instrument is intended to be a rapid, sensitive, and straightforward method of characterizing waste packages before they are sent to the WMA. In order to be assured of meaningful results, a series of validation tests has been performed using the instrument, and its performance has been compared to that of other characterization methods.

The manufacturer has provided an Operator Manual for the Waste Assay System. It includes a description of the components and their functions, an outline of the theory behind the calculations performed by the software, and detailed instructions for operating the WAS. Appendices provide supplementary information on energy and efficiency calibrations. The appendix entitled “Calibration Confirmation Test,” also published in [2], can be considered a component of the overall commissioning process. The confirmation test reports the results of measurements of a Eu-152 standard gamma source placed in reproducible positions within a plywood-filled B-25 container. The type of plywood was selected to provide a matrix density of approximately 0.5 g/cm^3, similar to typical densities of the wastes to be assayed. The results show the expected dependence of the signal intensity on the position of the source as well as the expected variation in signal attenuation with gamma energy.

Other commissioning information provided by the manufacturer includes dimensional and performance specifications of the two germanium detectors used for the gamma spectrometry measurements. Since the original commissioning of the instrument by the manufacturer, additional information has been provided and included as part of the paper and electronic documentation of the system. Based on AECL feedback, an updated Operator Manual has also been provided, with more details on both the theory and the operation of the WAS.

Background Checks

As part of the validation process, background spectra have been collected with the WAS in several different configurations. Count times were a minimum of 16 hours, to provide good sensitivity. For some spectra the platform was empty, for some it held an empty metal B-1000 box, and for some the test box containing the plywood was in place. The measurements were made after removing all movable sources of gamma activity, which included an active vacuum cleaner and any active boxes in the vicinity. The spectra were processed using GammaVision's ENV32 analysis module, in the same way as regular box spectra.

Several isotopes were detected, including K-40, some decay daughters of U-238 and Th-232, and the key contaminant at WL, namely Cs-137. The results are presented in Table 1. As expected, the lowest count rates are for the plywood-containing box, with the empty B-1000 box resulting in intermediate count rates, and the highest count rates occurring when no box is present on the load cell. This confirms our expectations that the empty box and the plywood-filled box are acting as gamma shields, reducing the exposure of the detectors to more distant sources of radioactivity. Even for the highest energy examined, namely the 1461-keV gamma ray of K-40, the empty B-1000 box and the plywood box provide significant attenuation of the background signal (plywood more than empty box). This pattern is consistent in both detectors, and the effect of the box is evident for the individual isotopes as well as for the overall count rates summed over the entire spectrum.

For all nuclides, the left detector (which faces the center of the room) shows consistently higher count rates than the right detector (which faces the outer wall). The difference is less than a factor of two for U and Th series nuclides, somewhat greater for K-40, and more than an order of magnitude for Cs-137. The efficiency of the detectors may have some effect, but the main contributor to the activity differences is what the detectors are aimed at. There is measurable Cs-137 contamination in the field of view of the left detector – most likely from the B-1000 waste compactor 10 m away.

What do these measurements indicate as far as routine Box Counter measurements go? Since we are assaying full boxes, the full (i.e. plywood-containing) box is the best one to use for Extended Background measurements. Up to this point, subtracting the extended background (using the spectrum-stripping option of GammaVision) does not make a significant difference to the assay results, but it may eventually do so, as the compactor becomes more contaminated over time.

Averaging Method

The WAS has three options for collecting gamma spectra of a waste container. All involve simultaneous measurements from both sides of the container, using the two gamma detectors of the instrument. In one case, the detectors are positioned far enough away that the entire box is within each detector's field of view; the measurement is known as a 1-position scan. For the second option, the detectors are positioned closer, so each one views approximately one quarter of the container in each position. Therefore four sequential measurements are required, with the box and detectors repositioned automatically between each measurement. This is termed a 4-position scan. Finally, the 6-position scan option has the detectors even closer to the container, so their fields of view are each constrained to approximately one-sixth of the container; this measurement option provides the highest sensitivity and the best evaluation of container homogeneity, but takes longer to complete.

It is important to verify that for a homogeneous container (uniform distribution of both radionuclides and matrix material) the three measurement options all produce the same results, when measurement uncertainty is taken into account. For the 4-position and 6-position scans, the Box Counter software is required to produce an average, or best-estimate value, of the total activity in the box. Extended discussions with the manufacturer to select the best averaging option took place in 2012, with eventual agreement on a method based on weighted arithmetic means.

To test the validity of the averaging method, several waste containers were measured using two or all three of the scanning options. Sample results for one box are shown in Table 3. For this particular box, spectra taken from opposite sides indicated that the waste inside was not distributed homogeneously, thus presenting a challenge to the averaging protocol. The results for the 1-position and 6-position scans may differ by a factor of two or more, but they are always within an order of magnitude of each other. Investigations of more robust averaging algorithms are continuing.

Scaling Factors for FM

In determining the radionuclide inventory of waste containers, it is recognized that not all radionuclides emit gamma rays. Since the WAS detectors are sensitive only to gamma rays, an estimate is required of radionuclides that are not gamma emitters, or are difficult to measure for some other reason. An internationally recognized approach is to characterize waste streams and establish empirical correlations between difficult-to-measure radionuclides and others that are easier to measure [4]. This approach is common practice in the nuclear industry to avoid the need for detailed analysis on a routine basis and has been incorporated into the WAS software.

The scaling factors are ratios based on key radionuclides that are both straightforward to measure and indicative of the behaviour of the difficult-to-measure ones. For example, the two fission products Cs-137 and Sr-90 are produced in similar quantities, and have similar half-lives. Using measurements of Cs-137 activity to estimate the activity of Sr-90 (a pure beta emitter) is justified once enough characterization information has been compiled to establish their most likely ratio in wastes (and, if possible, the degree of uncertainty in that ratio). Similarly, the radioactivity of the activation product Co-60 can be used to calculate the activity of many other activation products, based on empirical scaling factors. The gamma-emitting actinide Am-241 can be used to estimate the concentration of other actinides, specifically the various isotopes of plutonium, if the age and source of the actinides is known. Table 2 lists an example of the ratios used to calculate amounts of difficult-to-measure nuclides when the amounts of these three key marker radionuclides are known. The WAS software obtains the scaling-factor values from a user-supplied text file. If a key marker radionuclide is identified above the detection limit, the software then calculates estimates of values based on the appropriate scaling factors and includes them in the final report for analyzed containers, where the estimates are marked with the indication "–SF" to distinguish them from results based on direct detection of gamma ray peaks. If the marker radionuclide is not identified above the detection limit, the associated scaling factors are not applied.

VALIDATION STATUS

Comparison with ISOCS

At WL, portable gamma-ray spectrometers have been in use for waste characterization for several years, supporting both decommissioning and operations activities [5] [6]. The chief operator has gained considerable experience with measurements of various container types including B-25 and B-1000 containers. As part of the validation process, the radionuclide content of several containers has been determined using both the WAS and a portable spectrometer. The latter instrument is known as ISOCS, for In-Situ Object Counting System, manufactured by Canberra, Inc. It incorporates modelling calculations as part of the analysis process, using parameters of the gamma detector established at the factory, point-source calibrations, user-determined characteristics of the waste package, and the relative positions of waste and detector [7]. Measurements with the ISOCS instrument at WL are normally set up to collect a gamma spectrum with the entire side of the container in the detector's view, and then to sum the results from all four sides, or else from the two long sides, if time is limited.

The last column of Table 3 presents ISOCS results for the same box that was analyzed using the WAS. The comparison to ISOCS results shows significant differences, again up to an order of magnitude. Agreement is better with the WAS 1-position scan than with the 6-position scan, indicating that perhaps the initial averaging protocol is introducing a bias.

Remaining steps

With some additional effort, the issue of averaging of multiple gamma measurements should be resolved. Data obtained with 4-position scans will be compared to the existing data to see whether that approach is also subject to the same uncertainty as the original 6-position approach. Once the averaging method has been optimized, another round of comparisons with ISOCS portable gamma spectrometry will be undertaken. Further confirmatory information could be obtained if a box known to be homogeneous is available; e.g., one that contains uncontaminated soil, or well-mixed soil. A sample from that box could be analyzed in the traditional way, using a laboratory gamma spectrometry system, to provide a link to standard analytical methods and results.

CONCLUSION

The Waste Assay System is nearing routine use for confirmatory measurements of radiological inventory and characterization of waste containers destined for interim storage. Validation of its operating characteristics provides confidence that the results it generates will remain useful during the entire span of decommissioning for AECL's WL.

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Table 1. Box Counter Extended Backgrounds -- 16-hour Counts

Nuclide:	**Pb-212**	**Pb-214**	**Tl-208**	**Bi-214**	**Cs-137**	**Ac-228**	**K-40**	**Pb-212**	**Pb-214**	**Tl-208**	**Bi-214**	**Cs-137**	**Ac-228**	**K-40**
Energy (keV):	238	352	583	609	661	911	1461	238	352	583	609	661	911	1461
	Left detector (counts per second)							**Right detector (counts per second)**						
Plywood box	0.01	0.01	<0.01	0.01	0.01	<0.01	0.02	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.02
Empty B-1000	0.02	0.02	0.01	0.02	0.10	0.01	0.08	0.01	0.01	<0.01	0.01	<0.01	<0.01	0.03
No box	0.05	0.03	0.02	0.03	0.17	0.02	0.11	0.01	0.03	0.01	0.03	0.003	0.01	0.05

Table 2. Example of Box Counter Scaling Factors

Marker Radionuclide	DTM[a] Radionuclide	Activity Ratio
Cs-137	Sr-90	6.71E-01
Cs-137	Tc-99	1.67E-04
Cs-137	Sb-125	1.16E-02
Cs-137	Cs-134	2.53E-02
Cs-137	Pm-147	2.64E-01
Cs-137	Sm-151	2.94E-03
Cs-137	Eu-154	2.43E-02
Cs-137	Eu-155	1.03E-02
Am-241	Th-234	1.10E-03
Am-241	U-233	6.96E-09
Am-241	U-234	8.96E-04
Am-241	U-235	1.45E-05
Am-241	U-236	1.66E-04
Am-241	U-237	1.21E-03
Am-241	U-238	1.10E-03
Am-241	Np-237	9.10E-05
Am-241	Pu-238	2.74E-01
Am-241	Pu-239	5.73E-01
Am-241	Pu-240	7.84E-01
Am-241	Pu-241	4.84E+01
Am-241	Am-243	1.75E-03
Am-241	Cm-244	4.04E-02
Co-60	C-14	2.82E-03
Co-60	Fe-55	2.32E-01
Co-60	Ni-59	6.73E-05
Co-60	Ni-63	8.88E-03
Co-60	Zr-93	6.28E-04
Co-60	Nb-93m	2.46E-04
Co-60	Nb-94	1.55E-04

a. Note: DTM denotes “difficult to measure”

Table 3. Comparison of Box Counter Results (Two Scanning Options) with ISOCS Results

Nuclide	6-position scan	1-position scan	ISOCS results
K-40	0.091	0.111	<0.43
Co-60	0.019	0.019	<0.029
Cs-137	56.6	65.2	78.3
Eu-154	0.55	0.61	0.62
Bi-214	0.07	0.07	0.15
Am-241	478	3320	2090

Note: Activity values for nuclides in Box 368 are expressed as MBq.