14-A-368-INMM

RadSearch MEASUREMENTS FOR THE QUANTIFICATION OF CESIUM AND COBALT IN LEGACY SODIUM LOOP SECTION WASTE

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ABSTRACT

Sodium loop sections were irradiated in several nuclear reactors at the Idaho National Laboratory (INL) in the 1970's and 1980's in order to study the effects of high temperature and off normal conditions. The remaining waste was packaged for disposal in dual layer steel waste containers at INL. These containers have recently been retrieved by CH2M-WG Idaho, LLC. (CWI) for processing to separate the reactive metals from the matrix and then package the waste for final disposal. Historical data on the waste containers indicated a significant quantity of cesium-137 in the various containers and this information has been used to calculate the expected dose-rate and current Cs-137 content. The expectation was that other nuclides would have decayed away in the roughly 40 years since the packages were loaded. The ANTECH RadSearch G3050 Gamma Camera was used to take measurements of the waste containers in a number of configurations in order to establish how the canisters could be assayed prior to processing. Two canisters were selected for measurement, which are currently stored in concrete shielded containers. Three measurement configurations were employed including: indirect measurement through the concrete shield, indirect measurement through a carbon steel shield bell used to transfer such containers, and direct measurement of the container in open air. The first two measurements showed roughly the location of the container within the shield; however, the spectra were degraded so nuclide identification was not possible. Measurement of the container in open air provided data on the dose-rates and the nuclides present. Contact dose-rates on the measured containers were typically about 2 Sv/h (200 rem/h) and in one case the dose-rate was roughly a factor of 100 higher than expected from the recorded Cs-137 content. A goal of the measurement campaign was to investigate which radionuclides were contributing to the high dose-rate and to gain information on the spatial distribution of the radionuclides. Results of the measurements showed that the Cs-137 content was as expected but that in some cases, a significant quantity of Co-60 was also present. The measurements also showed that the Cs-137 spatial distribution was not uniform.

INTRODUCTION

The RadSearch G3050 Gamma Camera has been employed at the Idaho National Laboratory (INL) to measure the activity of dual layer steel waste containers used to store the irradiated contents of sodium loop sections. The purpose of the preliminary investigation of two waste containers (or cans) was to establish a method to determine the waste container activity based on gamma ray measurements and Monte Carlo simulations.

The RadSearch Gamma Camera [1,2] consists of a sensitive and highly collimated LaBr3 scintillation detector with an optical (video) camera with controllable zoom and focus and a laser range finder for distance measurement. The detector is housed in an effective tungsten shield, which provides a shielding ratio of 50:1 in the forward direction and 10:1 on the sides and to the rear for incident 1,500 keV gamma rays. The detector head is mounted on a pan/tilt mechanism with a range of motion of 360 degrees (pan) and +/- 90 degrees (tilt). For the present measurement campaign the detector head with pan/tilt is mounted on a mobile stand for ease of positioning. A single combined power and communication cable of 60 metres connects the detector head to an operator station consisting of a small power supply box connected to 110 or 230 VAC and a notebook computer. Operation of the detector head with all of its measuring and analysis functions is controlled from the notebook computer over Ethernet.

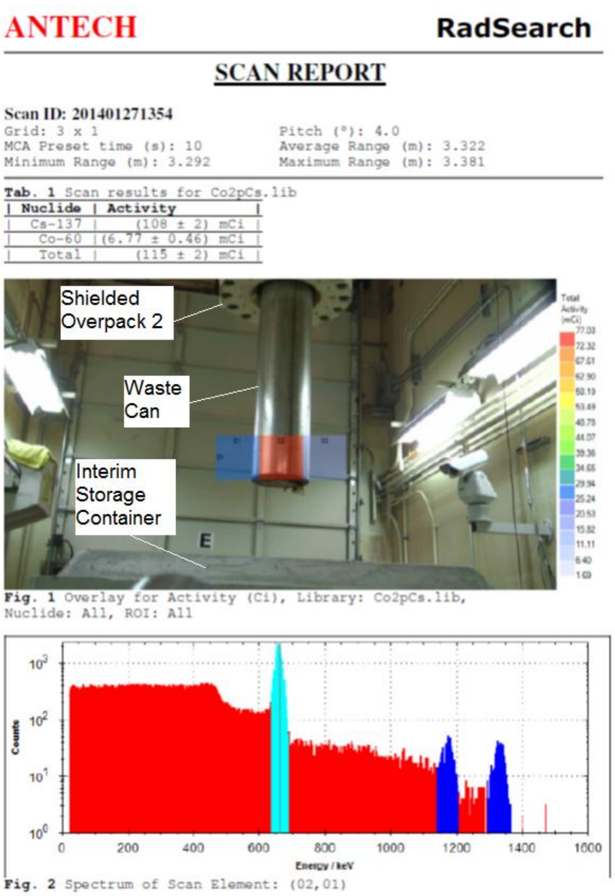
Figure 1. The photograph shows the RadSearch detector head with collimator barrel and pan-tilt mechanism on a mobile stand. The cylindrical object in the photo is a Shielded Overpack 2 (SO2), which is used when a waste container is transferred.

The high activity waste containers, which are the subject of the investigation, are of unknown density. It is expected, however, that most of the activity in the containers is due to the presence of Cs-137 with lesser amount of Co-60. Ordinarily, four waste containers are stored in a shielded Interim Storage Container (ISC), which is a concrete block with four re-entrant tubes with overall dimensions of 80 x 80 x 100 inches (203.2 x 203.2 x 254 cm). The task of transferring a waste container is performed using a Shielded Overpack 2 (SO2), which is a steel cylinder 32 inches in diameter and 75 inches long with a wall thickness of 9 inches of steel (81.3 cm OD x 190.5 cm L with wall thickness 22.9 cm).

In order to determine the activity of the waste containers the measured RadSearch count rate data is combined with the results of a Monte Carlo simulation, which is based on assumptions of homogeneity. The cylindrical waste containers are measured in vertical segments and the density is determined from the differential peak absorption of the two gamma ray peaks of Co-60 at energies of 1173.2 and 1332.5 keV, respectively. An MCNP simulation is used to determine the matrix density for each segment of each container from the peak ratio data derived from count rate measurements. Using the density for each segment of each container, another MCNP simulation is used to calculate the detection efficiency and hence the activity for each waste container segment assuming that both attenuation and source distribution is uniformly distributed (homogeneous) over the volume of each waste container. RadSearch and a Shielded Overpack 2 container are shown in Figure 1.

MEASUREMENTS

The significant activity in an unshielded waste container produces a very high dose-rate at the detector measurement position. To reduce the count rate from the waste container, RadSearch is equipped with a steel filter plug inserted into the end of the collimator barrel. The steel plug has a length of 51.6 mm, resulting in a 5% transmission of the 662 keV photons from Cs-137. This plug has the advantage of significantly suppressing the Compton scattered photons emanating from the inner volume of the waste containers. This keeps the spectrum integral count rate to a manageable level so that the counting electronics are not saturated.


ANTECH RadSearch

SCAN REPORT

Scan ID: 201401271354

Grid: 3 x 1 Pitch (°): 4.0
MCA Preset time (s): 10 Average Range (m): 3.322
Minimum Range (m): 3.292 Maximum Range (m): 3.381

Tab. 1 Scan results for Co2pCs.lib

Nuclide	Activity
Cs-137	(108 ± 2) mCi
Co-60	(6.77 ± 0.46) mCi
Total	(115 ± 2) mCi

Fig. 1 Overlay for Activity (Ci), Library: Co2pCs.lib, Nuclide: All, ROI: All

Fig. 2 Spectrum of Scan Element: (02,01)

Figure 2. The displayed image is a Scan Report from the RadSearch software showing a video image of the measured waste container (Fig. 1) and the gamma ray spectrum for the central (red) scan element (Fig. 2). Note that gamma ray peaks from Cs-137 and Co-60 are present in the spectrum. The reported activity is incorrect as it is assumed to be from a surface source.

Currently, the RadSearch analysis software is designed for the measurement of the activity of a point or area source located on the surface of an object. The distance to the surface is measured with a laser rangefinder and the activity is calculated using the known efficiency for specific gamma ray energies. The instrument is calibrated by performing a scan of a calibrated and traceable gamma point source placed at a fixed distance. To determine the activity of a volume source the raw measurement results, i.e. the net peak count rates, must be post-processed to correct for the source distribution and the source self-shielding by the waste matrix inside the waste containers. This calculation is performed using a Monte Carlo simulation.

After initial tests through the ISC and SO2 shields, the measurement of each waste container was performed with the SO2 shielding partially removed to expose part of the waste container. In order to limit the exposure of personnel during the measurements the waste container was partially shielded by the ISC at the bottom and the SO2 at the top, as is shown in Figure 2, (see Note below). The SO2 was kept at a fixed height, i.e. suspended by a crane, whereas the waste container was positioned at various heights using a hoist, which is incorporated into the SO2. An attempt was made to keep the vertical translation of the waste container equal to the size of one scan-element (a square area on the screen the size of the scan pitch, i.e. 4°). As a result 9 or 10 vertical segments are required to cover the entire height of the waste container. (Note: Within Figure 2, which is itself a RadSearch Report, the video image and the spectrum are internally labelled Fig. 1 and Fig. 2).

Measured data for the two test waste containers No. 007 and No. 016 are tabulated in Tables 1 and 2. The net peak count rates are provided by the RadSearch software, which allows the user to select the width of the Region Of Interest (ROI) used to calculate the net peak area. In this analysis the width of the ROI is equal to 2.5 times the Full Width Half Maximum (FWHM) for a specific gamma ray peak. This is considered optimum for the medium count rates encountered in these measurements.

The presence of Co-60 in all the segments permits the determination of the density of the waste matrix by making use of the slightly different mass attenuation coefficients for the two Co-60 peaks. The peak ratio, R, is given by:

$$R = \frac{C_{1173}}{C_{1333}} \quad (1),$$

where C_{1173} and C_{1333} are respectively the net count rates for the Co-60 peaks at 1173 and 1333 keV.

Segment	ScanID	Net Peak Count Rates (s^{-1})						Peak Ratio *R*	
		Cs-137 662 keV		Co-60 1173 keV		Co-60 1333 keV			
		Value	Error	Value	Error	Value	Error	Value	Error
1	201401271354	1527	27	33	5	39	4	0.86	0.18
2	201401271353	1281	26	36	6	45	4	0.81	0.17
3	201401271352	38	14	185	12	207	10	0.89	0.07
4	201401271350	25	12	134	10	162	9	0.83	0.08
5	201401271348	39	11	124	9	133	8	0.94	0.09
6	201401271347	26	11	149	10	167	9	0.89	0.08
7	201401271346	4	9	115	9	131	8	0.87	0.09
8	201401271345	0	0	85	8	102	7	0.83	0.10
9	201401271344	4	5	30	4	38	4	0.81	0.16
10	201401271342	0	1	0	1	0	1		

Table 1. Measured count rates for the central scan-element for various vertical segments of Waste Container No. 007. The peak ratio, *R*, is also given. The errors correspond to 2σ. Segment 1 corresponds to the bottom of the waste container.

Segment	Scan ID	Net Peak Count Rates (s^{-1})						Peak Ratio *R*	
		Cs-137 662 keV		Co-60 1173 keV		Co-60 1333 keV			
		Value	Error	Value	Error	Value	Error	Value	Error
1	201401271542	4.1	7.5	35.9	6.1	58.6	5.1	0.61	0.14
2	201401271541	9.9	13.5	259.0	13.2	283.1	11.2	0.91	0.06
3	201401271540	10.9	18.7	620.9	19.7	662.6	16.9	0.94	0.04
4	201401271539	13.4	17.1	445.5	17.1	493.2	14.8	0.90	0.05
5	201401271538	16.9	19.9	611.5	20.3	683.6	17.4	0.89	0.04
6	201401271537	183.2	20.2	513.1	18.7	590.7	16.7	0.87	0.04
7	201401271536	286.2	17.4	284.6	14.1	340.5	12.2	0.84	0.05
8	201401271534	66.4	8.4	66.9	6.5	65.8	5.3	1.02	0.13
9	201401271533	0.9	2.0	1.6	1.3	1.9	1.0	0.84	0.86

Table 2. Measured count rates for the central scan-element for various vertical segments of Waste Container No. 016. The peak ratio, *R*, is also given. The errors correspond to 2σ. Segment 1 corresponds to the bottom of the waste container.

As can be seen in Tables 1 and 2, the errors in the peak ratio, *R*, are quite large. This is because they depend on the error of both the 1173 and 1333 keV net peak count rates. Unfortunately, this large error results in a loss of precision in the determination of the matrix densities.

MONTE CARLO MCNP SIMULATIONS

The MCNP4C2 Monte Carlo code [3] is used to determine the matrix density for a measured peak ratio. In fact the ratio in the observed net peak count rates is equal to the ratio in the efficiencies. The quality of the model is of fundamental importance in obtaining valid results: the MCNP model of RadSearch has been validated by numerous experimental results [2]. The geometry of the model is shown in Figures 2 (in the video image) and 3 and is based on a detailed drawing provided by INL and on the range and tilt of RadSearch in relation to the waste container.

The main uncertainty in the simulation of the waste container measurements is the composition of the waste matrix. In this study a waste matrix of iron (Fe) is used, but as can be seen in Table 3, the results will be very similar if a sodium matrix (Na) were used instead. The mass attention values between 600 and 1500 keV are very similar for these two elements. The difference between the values for the two Co-60 peaks, which determines the peak ratio, *R*, is virtually the same for the two elements. The presence of hydrogenous material in the waste would have only a minor impact on the peak ratio because of the lower density of hydrogen, even though hydrogen has a quite different mass attenuation characteristics.

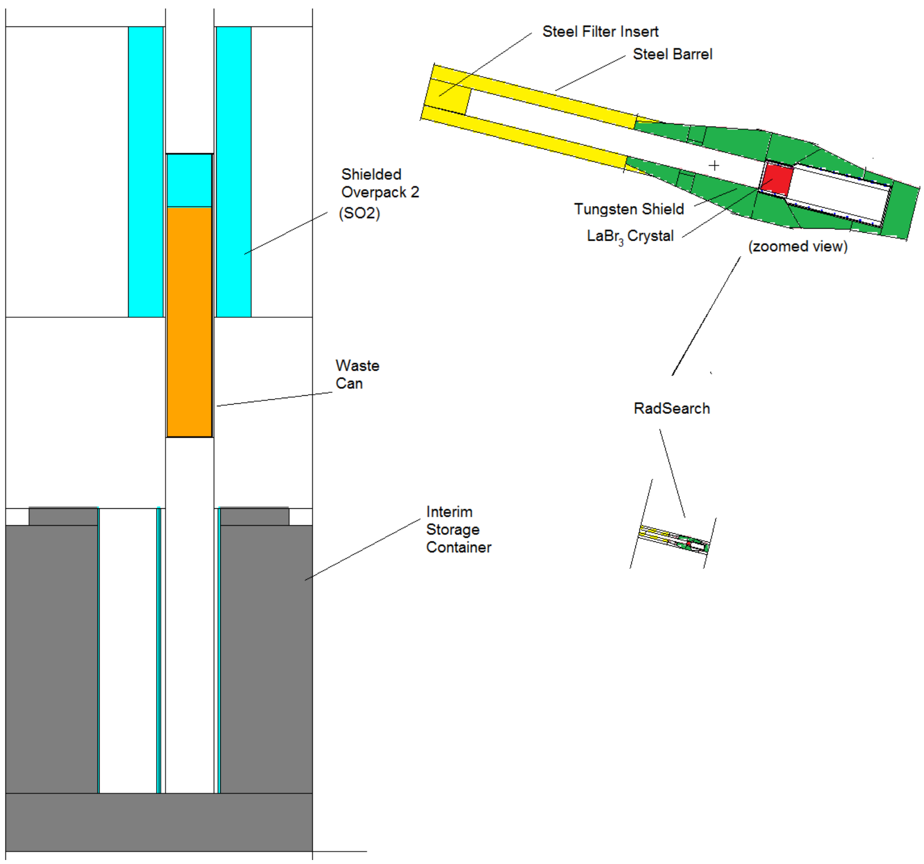


Figure 3. View of a vertical section through the centre of the waste container or can (from the MCNP Model).

Photon Energy (keV)	$(\mu/\rho)_{Na}$ ($cm^2.g^{-1}$)	$(\mu/\rho)_{Fe}$ ($cm^2.g^{-1}$)	Relative
662	0.074437	0.073942	1.006694
1173	0.056524	0.05548	1.018818
1333	0.052936	0.051959	1.018803
Δ	0.936533	0.936525	1.000009

Table 3. Mass attenuation coefficients (μ/ρ) in $cm^2.g^{-1}$ for sodium (Na) and iron (Fe) for photon energies of interest are shown in Table 3. The values were obtained by linear interpolation from data by Hubbell and Seltzer [4]. The values in the column with heading Relative, is obtained by dividing the values for Na by those for Fe. The values in the row with Δ in the first column are the difference in the mass attenuation for the two Co-60 peaks.

MEASUREMENT RESULTS

The measured activity, *A* (in Bq), is given by the following equation:

$$A = \frac{C}{0.01\ \varepsilon\ I_\gamma} \quad (2),$$

where *C* is the measured net count rate (in s^{-1}), ε, is the detection efficiency and, $I\gamma$, is the yield per disintegration for a specific gamma energy in percent (also called the Branching Ratio).

Nuclide	Peak Energy (keV)	I_γ (%)
Cs-137	661.657	85.1
Co-60	1173.237	99.9736
Co-60	1332.501	99.9856

Table 4. Branching Ratios for peaks of interest.

The method to calculate the final results is as follows:

- Determine the peak ratio, *R*, from the measured net peak count rates;
- With MCNP simulations find the density, ρ, of the waste matrix that gives the same peak ratio, *R*;
- Calculate with MCNP the detection efficiency, ε, of the gamma rays of interest for the previously obtained density, ρ;
- Calculate the activity with equation (2).

The results for the radionuclide activity in Waste Containers 007 and 016 are given in Tables 5 and 6 respectively. Note that the density values in bold were adjusted manually. Due to the large uncertainty in the peak ratio, *R*, some values of *R* would result in unrealistically large densities. The given values are based on the spectrum shapes. The errors for Waste Can 016 were smaller and no adjustments were required. The results are represented graphically in Figures 4 and 5.

Segment	Calculated Density ($g.cm^{-3}$)	MCNP Efficiency ($photon^{-1}$)			Calculated Activity (Ci)		
		Cs-137 @ 662 keV	Co-60 @ 1173 keV	Co-60 @ 1333 keV	Cs-137	Co-60 @ 1173 keV	Co-60 @ 1333 keV
1	**4.5**	8.40E-10	1.26E-09	1.41E-09	57.82	0.71	0.74
2	**4.5**	8.40E-10	1.26E-09	1.41E-09	48.51	0.78	0.86
3	4.3	8.61E-10	1.32E-09	1.47E-09	1.39	3.79	3.80
4	**4.5**	8.40E-10	1.26E-09	1.41E-09	0.95	2.87	3.11
5	2.5	1.29E-09	2.17E-09	2.33E-09	0.95	1.54	1.54
6	4.3	8.58E-10	1.31E-09	1.46E-09	0.96	3.07	3.08
7	5.8	7.83E-10	1.07E-09	1.23E-09	0.17	2.90	2.90
8	**2.5**	1.32E-09	2.20E-09	2.36E-09	0.00	1.04	1.17
9	**2.5**	1.32E-09	2.20E-09	2.36E-09	0.10	0.37	0.43
10	**2.5**	1.32E-09	2.20E-09	2.36E-09	0.00	0.00	0.00
				Total	**110.85**	**17.07**	**17.63**

Table 5. Measurement results for the Activity in Waste Container 007 based on detection efficiencies (per starting photon) calculated with MCNP. The branching ratios in Table 4 have been used to calculate the activity with equation 2. The densities have been calculated by using the experimental peak ratios for the two Co-60 peaks and MCNP simulation results.

Segment	Calculated Density ($g.cm^{-3}$)	MCNP Efficiency ($photon^{-1}$)			Calculated Activity (Ci)		
		Cs-137 @ 662 keV	Co-60 @ 1173 keV	Co-60 @ 1333 keV	Cs-137	Co-60 @ 1173 keV	Co-60 @ 1333 keV
1	5.0	9.09E-10	1.43E-09	1.93E-09	0.14	0.68	0.82
2	1.9	2.16E-09	3.14E-09	3.43E-09	0.15	2.23	2.23
3	1.4	2.89E-09	4.04E-09	4.30E-09	0.12	4.15	4.16
4	2.1	1.92E-09	2.85E-09	3.15E-09	0.22	4.22	4.23
5	2.2	1.79E-09	2.67E-09	2.99E-09	0.30	6.19	6.18
6	2.6	1.50E-09	2.30E-09	2.66E-09	3.88	6.03	6.00
7	3.1	1.29E-09	2.00E-09	2.39E-09	7.05	3.85	3.85
8	0.0	7.83E-09	9.52E-09	1.00E-08	0.27	0.19	0.18
9	3.0	1.31E-09	2.04E-09	2.42E-09	0.02	0.02	0.02
				Total	**12.16**	**27.56**	**27.68**

Table 6. Measurement results for the activity in Waste Container 016 based on detection efficiencies (per starting photon) calculated with MCNP. The branching ratios in Table 4 have been used to calculate the activity with equation 2. The densities have been calculated by using the experimental peak ratios for the two Co-60 peaks and MCNP simulation results.

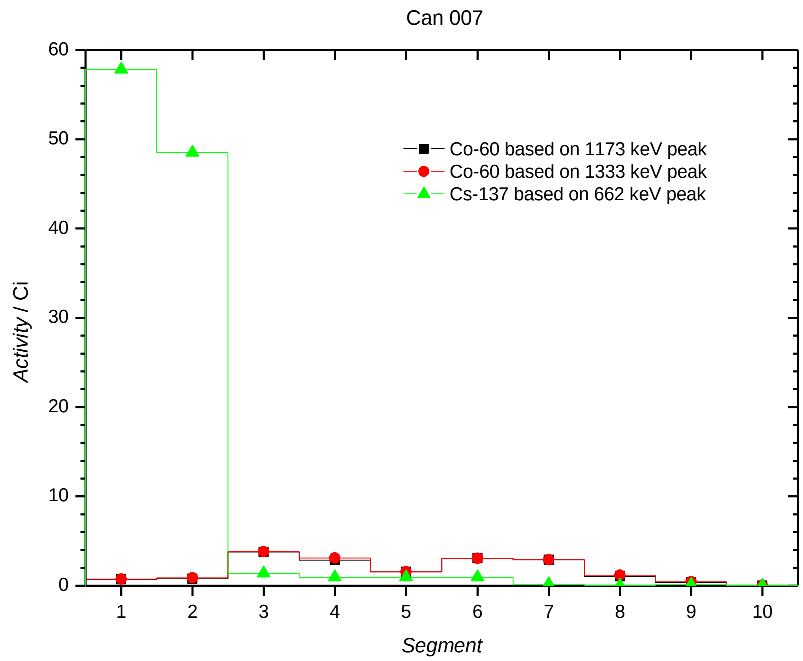


Figure 4. Measured activity (in units of Ci) as a function of the container segments for Waste Container 007.

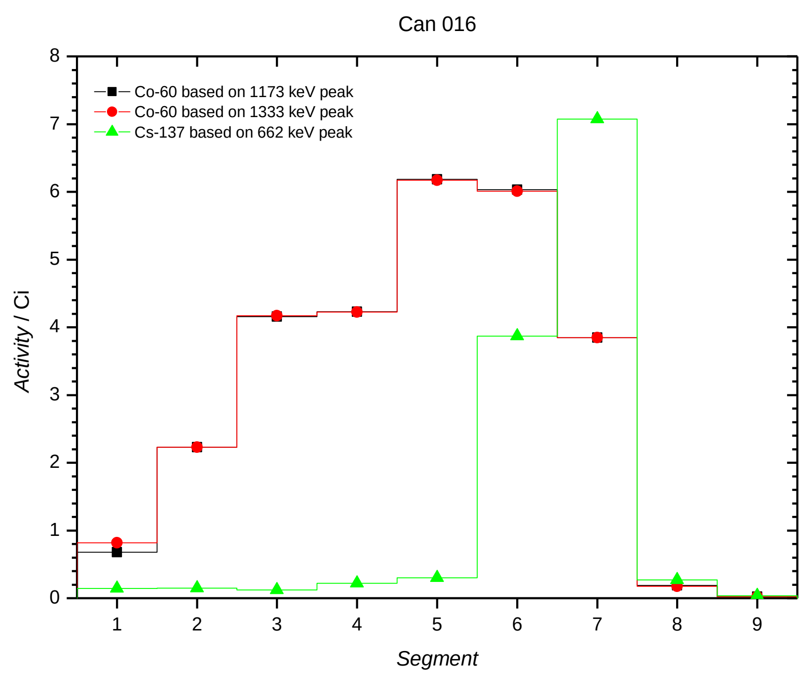


Figure 5. Measured activity (in units of Ci) as a function of the container segments for Waste Container 016.

A typical gamma ray spectrum from the RadSearch measurement is displayed in Figure 6, below. The measurement is of segment 6 of waste container 007. It is useful to compare this spectrum with Figure 4, which shows the relevant activities for both Cs-137 and Co-60 derived from the analysis for segment 6 of waste container 007.

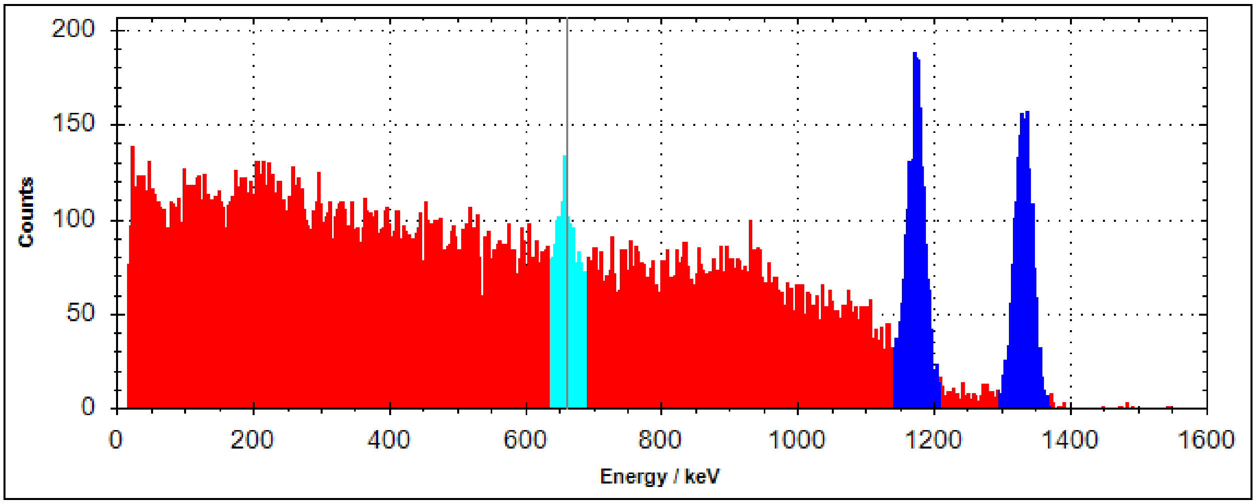


Figure 6. Gamma ray spectrum for the measurement of Waste Container 007, segment 6 (Scan ID= 201401271347), which contains gamma ray peaks for Cs-137 (661.64 keV) and Co-60 (1173.2 keV and 1332.5 keV).

CONCLUSIONS

By employing a combination of gamma ray spectroscopic measurements and MCNP Monte Carlo simulations the density and radionuclide activity distribution of high activity waste containers have been determined. The approach is dependent on the dual assumptions of matrix homogeneity and uniformity of radionuclide distribution. The method is applicable wherever a multiline gamma ray emitter is present (such as Co-60), so that differential attenuation may be employed to determine the density of the unknown sample matrix.

RadSearch allowed the containers to be measured in vertical segments, each segment with potentially a different density. As a result variations in both density and radionuclide distribution were determined. A higher than expected distribution of Co-60 was measured in waste container 016 as displayed in Figure 5.

The preliminary investigation reported in this paper has established an effective method and a measurement and analysis protocol to determine the waste container activity and radionuclide distribution that can be employed for the analysis of the remaining sodium loop section waste containers.

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