Screening and Spectral Summing of LANL Empty Waste Drums – 13226

Kathleen M. Gruetzmacher (kgruetzmacher@lanl.gov)*, Roland M. Bustos (rbustos@lanl.gov)*, Scott G. Ferran (ferrans@lanl.gov)*, Lucas E. Gallegos (lucas@lanl.gov)*, and Randy P. Lucero (rlucero@pscnda.com)**

*Los Alamos National Laboratory, Los Alamos, New Mexico 87545
**Pajarito Scientific Corporation, Santa Fe, New Mexico 87505

ABSTRACT

Empty 55-gallon drums that formerly held transuranic (TRU) waste (often over-packed in 85-gallon drums) are generated at LANL and require radiological characterization for disposition. These drums are typically measured and analyzed individually using high purity germanium (HPGe) gamma detectors. This approach can be resource and time intensive. For a project requiring several hundred drums to be characterized in a short time frame, an alternative approach was developed. The approach utilizes a combination of field screening and spectral summing that was required to be technically defensible and meet the Nevada Nuclear Security Site (NNSS) Waste Acceptance Criteria (WAC). In the screening phase of the operation, the drums were counted for 300 seconds (compared to 600 seconds for the typical approach) and checked against Low Level (LL)/TRU thresholds established for each drum configuration and detector. Multiple TRU nuclides and multiple gamma rays for each nuclide were evaluated using an automated spreadsheet utility that can process data from up to 42 drums at a time. Screening results were reviewed by an expert analyst to confirm the field LL/TRU determination. The spectral summing analysis technique combines spectral data (channel-by-channel) associated with a group of individual waste containers producing a composite spectrum. The grouped drums must meet specific similarity criteria. Another automated spreadsheet utility was used to spectral sum data from an unlimited number of similar drums grouped together. The composite spectrum represents a virtual combined drum for the group of drums and was analyzed using the SNAP™/Radioassay Data Sheet (RDS)/Batch Data Report (BDR) method. The activity results for a composite virtual drum were divided equally amongst the individual drums to generate characterization results for each individual drum in the group. An initial batch of approximately 500 drums were measured and analyzed in less than 2 months in 2011. A second batch of approximately 500 more drums were measured and analyzed during the following 2 1/2 months. Four different HPGe detectors were employed for the operation. The screening and spectral summing approach can reduce the overall measurement and analysis time required. However, developing the technical details and automation spreadsheets requires a significant amount of expert time prior to beginning field operations and must be considered in the overall project schedule. This approach has continued to be used for characterizing several hundred more empty drums in 2012 and is planned to continue in 2013.

INTRODUCTION

Empty 55-gallon drums that formerly held TRU waste are generated in ongoing drum repackaging operations at LANL. Once emptied, these drums may have residual radioactive contamination remaining inside the drum and so must be treated as radioactive waste themselves. The empty drums are typically of 55-gallon volume that may be over packed in an 85-gallon drum after being emptied. Historically, for radiological characterization, these empty drums have been measured and fully analyzed individually using HPGe gamma spectroscopy detectors. While measuring and fully analyzing each drum is completely effective at a high level of confidence towards determining the LL or TRU disposition route for the drums, it can be resource and time intensive. The protocol for this type of individual evaluation requires a measurement long enough to achieve good individual drum counting statistics (typically 600 seconds) and requires 2 qualified analysts (1 for review) for approximately 1 hour combined per drum. When a large number of drums need to be characterized in a short amount of time, this approach can be resource restricted. In 2011 several hundred drums had to be characterized within a couple months. Thus, an alternate characterization approach that is less time and resource intensive overall – but that still provides sufficient effectiveness and confidence in the LL/TRU disposition of the drums – was developed and implemented. The approach utilizes a combination of field screening and spectral summing and was applied to the population of empty drums.

REQUIREMENTS

For the 2011 empty drum characterization project, there were both technical and schedule requirements. The base technical requirements were: (1) each empty drum must be radiologically characterized such that a LL/TRU determination can be made; (2) the characterization method must meet the NNSS WAC for drums determined to be LL. The schedule requirement was that approximately 500 drums be characterized in approximately 2 months. Standard individual measurement and comprehensive analysis of each drum would generally require more than this allocated time to complete. Thus, an alternative characterization approach was necessary in order to achieve the project goals.

STANDARD METHODOLOGY

The standard approach to characterizing empty waste drums at LANL is to perform for each drum a 600 second non-destructive assay (NDA) measurement using a HPGe gamma detector followed by a comprehensive analysis of the resulting gamma spectrum using the SNAP™ analysis software and the RDS/BDR software. A 600 second measurement has been established to provide more than sufficient MDA results for LL/TRU determination in most cases. The SNAP™ analysis provides a modeled individual assay result that together with the RDS/BDR software provides activity values for detected radionuclides and MDA or scaled activity values for non-detected radionuclides of interest and establishes the LL/TRU disposition of the drum. This approach has been established to be technically rigorous and meet the NNSS WAC, however, it can be resource and time intensive – particularly for the SNAP™ analysis – as each analysis must be performed by a qualified analyst and reviewed by an independent qualified analyst. Figure 1 illustrates the main steps in the standard analysis methodology.

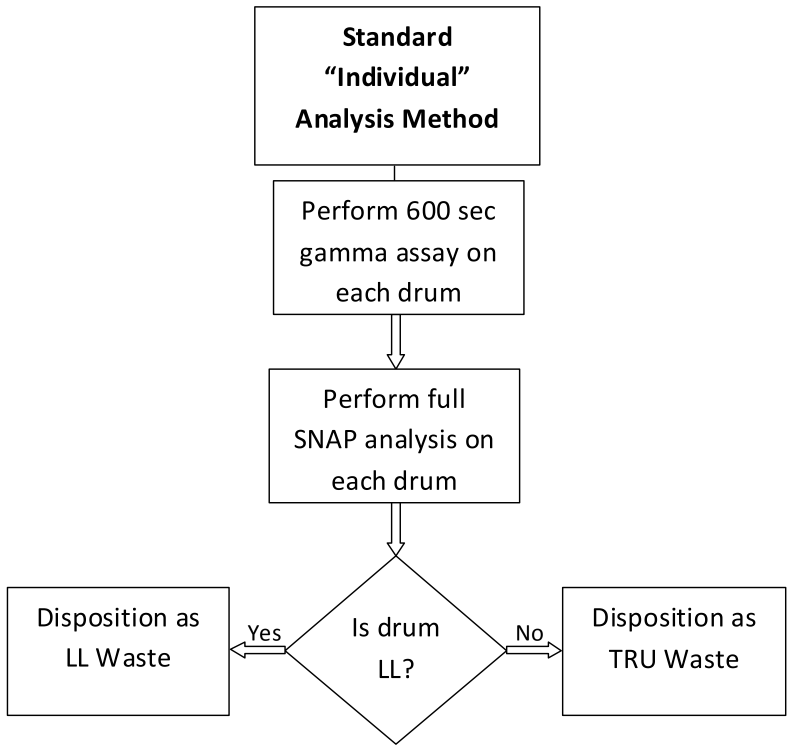

Fig. 1. Flowchart showing the main steps in the standard analysis methodology

SCREENING

For the screening phase of the operation each drum was assayed for a count time of 300 seconds with a HPGe detector – the count time was decreased from 600 seconds for a standard measurement to 300 seconds for the screening measurement. After numerous short drum measurements were completed (typically after a full measurement shift) a quantitative screen analysis was performed on the acquired spectra to determine whether each drum was LL or TRU. In the screen analysis the key TRU radionuclides present in LANL waste were evaluated: Am-241, Am-243, Cm-243, Pu-238, Pu-239 and Np-237. One or more gamma rays were evaluated for each nuclide and are listed in Table I.

Screening thresholds for peak net counts (upper limit) for each of the gamma rays listed in

TABLE I. Gamma rays evaluated for each radionuclide

Nuclide	Gamma Rays Evaluated
Am-241	59.5keV
	125.3keV
Am-243 and Cm-243	228.2keV
Pu-238	99.9keV
	152.6keV
Pu-239	129.3keV
	413.7keV
Np-237	311.9keV

Table I were established using the SNAP™ software. Two empty drum configurations for each detector were modeled to establish the thresholds based on the 3700Bq/g (100nCi/g) total TRU alpha activity concentration WIPP limit. The two configurations modeled were: 1) 55-Gallon drum with a poly liner; 2) 55-Gallon drum with a poly liner over packed in an 85-Gallon drum. The software models used were identical to the models that would be used in a standard individual drum analysis.

An automated spreadsheet utility was developed that reads up to 42 spectra and checks the peak net counts against the threshold values. This allowed a determination of the LL/TRU status of each drum in the batch in less than a minute batch processing time. If the same were performed manually, it would have taken much longer (probably at least 1/2 hour for 42 drums) and been more susceptible to error. An example of the output of the screening spreadsheet utility with the net counts displayed for the key Am-241 and Pu-239 gamma rays is shown in Figure 2. The thresholds were specific to each detector and drum configuration, so an automated spreadsheet utility was customized for each detector/drum configuration and drums were processed through the tailored spreadsheet.

The screening spreadsheet utility was quality checked before use. As a supplemental quality assurance step to confirm that drums that were field screened as LL were appropriately classified, a formal review of the screen results by a qualified gamma spectroscopy analyst was performed for every drum measured. This review was generally performed a day or more after the initial field screen and included viewing each drum spectrum and checking that the peak counts were translated correctly into the spreadsheet utility. Adjustments were made as necessary to the LL/TRU classification of the drum and the reviews were documented by the analyst with initial and date in a copy of the detector field log books. An example of a review record is shown in Figure 3.

ItemID	Date	Detector	Screen Result	LiveTime (s)	Real Time (s)	C60	C129
092911sam01	9/29/2011	Sam	LL	300	301.6	135.00	15.00
092911sam02	9/29/2011	Sam	TRU	300	309.8	58150.00	1138.00
092911sam02a	9/29/2011	Sam	TRU	300	309.8	58512.00	1079.00
092911sam03	9/29/2011	Sam	LL	300	301.9	81.00	1.00
092911sam04	9/29/2011	Sam	LL	300	301.7	202.00	30.00
092911sam05	9/29/2011	Sam	LL	300	302	3032.00	102.00
092911sam06	9/29/2011	Sam	LL	300	301.5	67.00	22.00
092911sam07	9/29/2011	Sam	LL	300	301.6	113.00	19.00
092911sam08	9/29/2011	Sam	LL	300	301.6	206.00	4.00
092911sam09	9/29/2011	Sam	LL	300	301.5	313.00	0.00
092911sam10	9/29/2011	Sam	LL	300	301.4	96.00	4.00
092911sam11	9/29/2011	Sam	LL	300	302	193.00	24.00
092911sam12	9/29/2011	Sam	LL	300	301.5	288.00	23.00
092911sam13	9/29/2011	Sam	LL	300	302.1	96.00	14.00
092911sam14	9/29/2011	Sam	LL	300	301.6	461.00	-14.00

Fig. 2. Example of the screening spreadsheet utility output

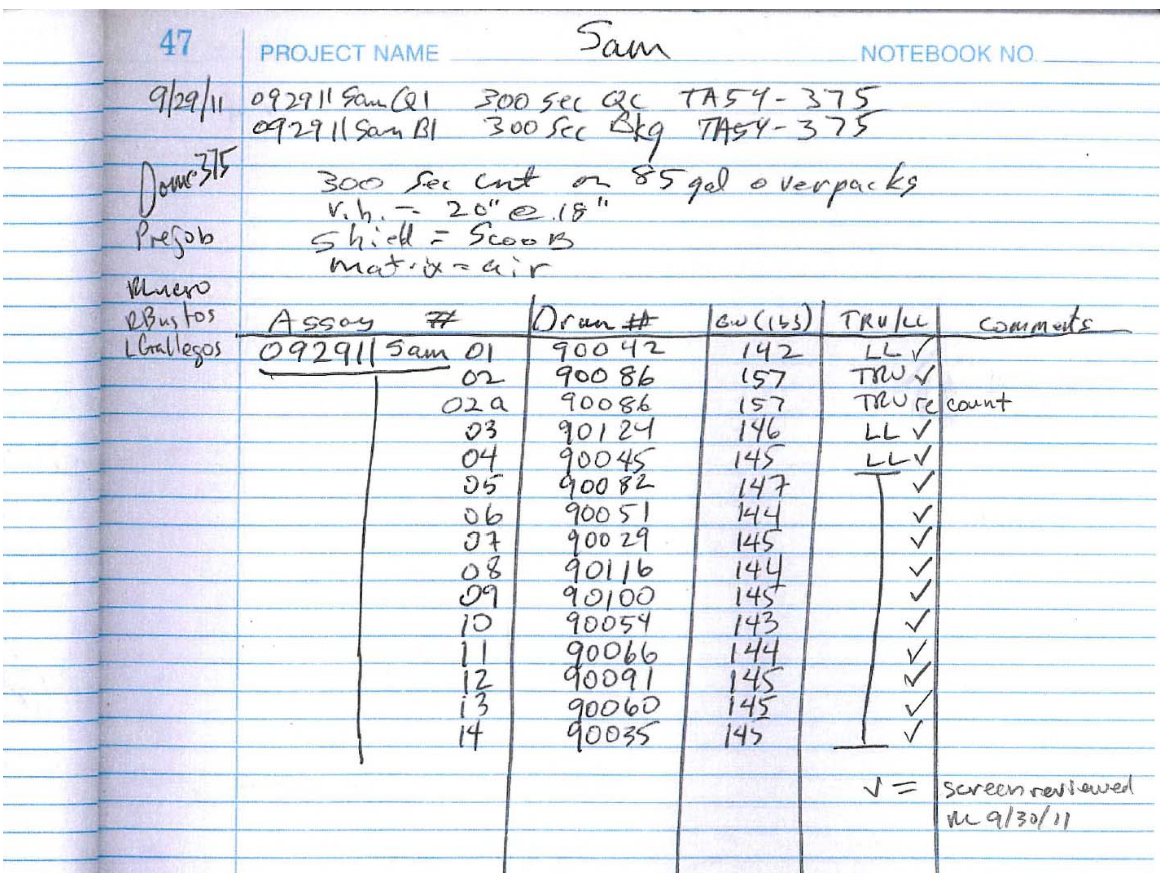

Fig. 3. Example of analyst post field screen review record

SPECTRAL SUMMING

After the screening phase of the operation, the drums that were classified as LL were analyzed together using spectral summing to generate detailed quantitative assay results. The spectral summing analysis technique combines spectral data associated with a group of individual waste containers, and provides a single radioassay result for the entire group. The method involves a channel-by-channel summation of the spectral data associated with a group of waste containers, which individually may have radionuclide activities that are below the MDA. The resulting spectrum, referred to as the composite spectrum, is then treated in the same way as any other spectrum acquired during the assay of an individual container. The spectral analysis and the NDA algorithms that were applied to the composite spectrum were the same as those that are applied to individual assays. The technique takes advantage of the improved counting statistics associated with the combined spectral data and in the present case greatly reduced the overall analysis time.

The spectral summing technique has been previously and successfully applied at LANL [1, 2] and has also been successfully applied and validated by Pajarito Scientific Corporation at the AMWTP at INEL [3].

Drum Grouping

Conceptually the composite spectrum may be viewed as a single assay event associated with a virtual waste container. The virtual container analysis is applied to all the individual drums in a group that make up a virtual waste container, so the containers must be sufficiently similar in configuration and waste type for the approach to be defensible. The mass of individual waste containers that were combined in the analysis defines the mass of a composite virtual waste container.

One key consideration for grouping the containers was the waste matrix type, e.g., air, debris, steel. For this operation all of the drums were empties, i.e., with a matrix of air, so all of the drums were grouped together in this category. Another key consideration was the size and wall thickness of the container. Two empty drum container configurations were present for this operation: 1) 55-Gallon drum with a poly liner; 2) 55-Gallon drum with a poly liner over packed in an 85-Gallon drum. These configurations were too different in container size and wall thickness to spectral sum together, so they were grouped separately for the spectral summing. Finally, detector characteristics such as resolution and intrinsic efficiency calibrations were not sufficiently equivalent that data acquired on different detectors could be spectral summed. Thus, drums were further grouped by the detector on which they were measured. In summary, the empty drums were grouped by matrix, container configuration and by detector for spectral summing.

Composite Analysis

Once drums were grouped according to the specified criteria, the spectra acquired separately for each individual drum in the group were summed together and a single SNAP™/RDS/BDR analysis was performed on the composite spectrum. An example of a composite spectrum (in royal dark blue) compared to a spectrum from an individual drum from the summed set (in turquoise) is shown in Figure 4. Since from two to several hundred drums may be grouped together for spectral summing, an automated spreadsheet utility was developed that can read an unlimited number (or only limited by the intrinsic spreadsheet limits) of individual drum spectra, sum them together in the acquisition software and create a composite spectrum that is compatible for analysis. Verification and validation (V&V) was performed and documented on the spectral summing spreadsheet utility [4].

The following parameters were applied to a composite virtual drum in the SNAP™ model:

1. A single physical model based on the shared drum type and configuration and detector configuration.

2. A 300 second count time (each drum was measured for 300 seconds).

3. The combined weight of the individual drums.

Using these analysis parameters provides an assay result that represents the total activity (Bq or Ci) in a composite virtual drum. The activity concentration (Bq/g; nCi/g in the RDS) for a composite virtual drum using the combined weights also represents the average activity concentration for each individual drum. Figure 5 shows an example of a RDS for a composite virtual drum analysis.

Drum Average Calculations

Once the composite spectrum results for a composite virtual drum were calculated using the SNAP™/RDS/BDR analysis method, activity results had to be determined and assigned for each individual drum in the spectral summed set. The activity results for a composite virtual drum represent the total activity in all the individual drums combined (for each radionuclide reported). So the activity result for each radionuclide identified in the composite analysis was divided equally amongst the individual drums in the summed set. For example, if the composite analysis resulted in 10GBq of activity for the radionuclide Pu-239, and 10 individual drum spectra were summed together to create the composite spectrum for a composite virtual drum, then each of the 10 individual drums would have an activity result for Pu-239 of 1GBq. The associated activity

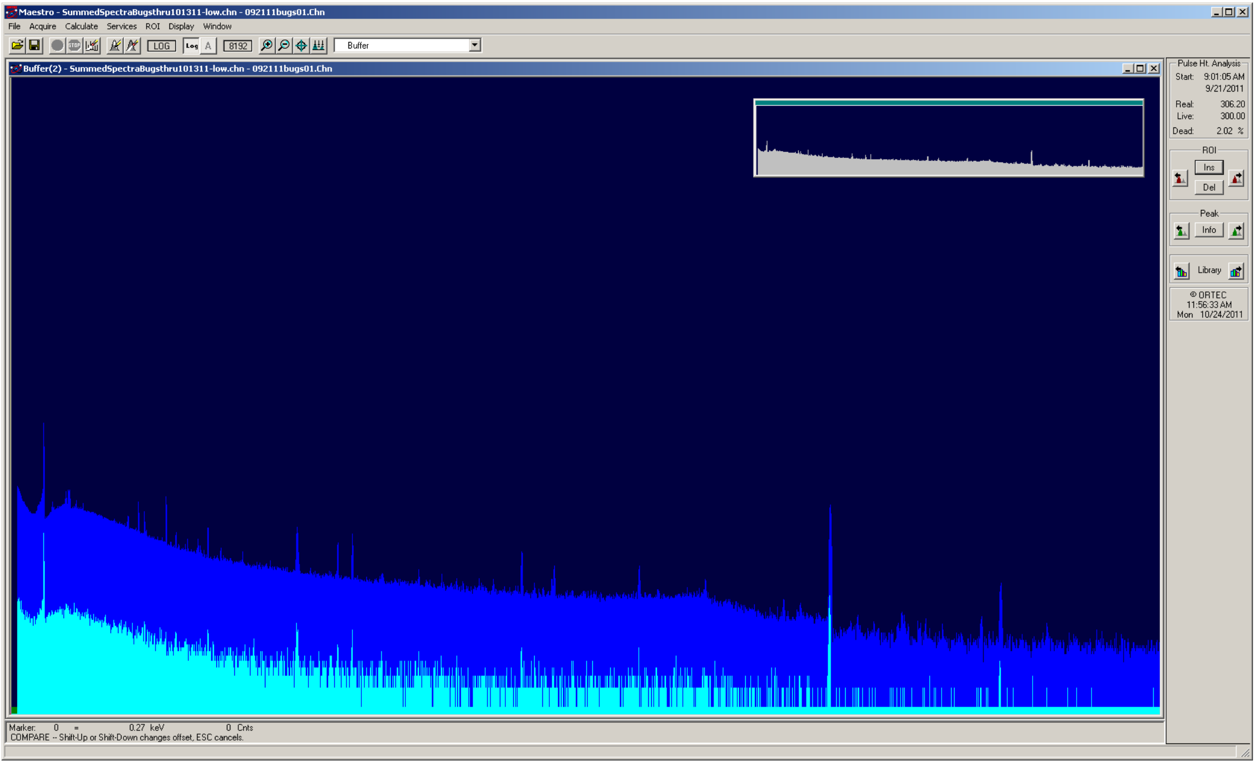

Fig. 4. Example of a composite spectrum compared against an individual drum spectrum

The activity concentration (Bq/g) for each of the individual drums was calculated (in units of nCi/g during calculations) using the average drum weight of all the individual drums in the set; thus, the activity concentration for a composite virtual drum was equal to the activity concentration for each individual drum. Figure 6 shows an example of a RDS for a drum from the spectral sum set used to generate the composite virtual drum RDS in Figure 5. Figure 7 illustrates the main steps in the alternative screening spectral summing analysis methodology.

DRUMS MEASURED AND PROCESSED

The initial drum set that prompted the development of the applied screening and spectral summing approach was comprised of approximately 500 empty drums. These drums were measured and screened between June 16, 2011 and July 21, 2011 on two detectors. The spectral summing analysis on the drums screened as LL was complete by August 2, 2011 and helped to achieve an important fiscal year end milestone. After that initial campaign, another approximately 500 drums were measured, screened and spectral summed as LLW before November 1, 2011. Four different HPGe detectors were used for the operation throughout the time period listed. In total approximately 150 drums were screened as TRU and analyzed using the standard method over several months following the screening measurements. Similar operations have continued in 2012 achieving the characterization of several hundred more empty drums and are planned to continue in 2013.

Radioassay Data Sheet

Assay Conclusion: LLW

MDAs Set with MT and Ref Isotope: PU52 (Pu239), U(NAT) (None)

Container ID:	Virtual Drum	*Site ID:*	LANL
Revision No:	0	*Procedure:*	Q2/SNAP
File Name:	[illegible]	*Software 1:*	SnapV1.13
Detector:	Minnie	*Software 2:*	Peak Doctor v1.0
Assay Date and Time:	10/19/2011 0:00	*Assay Method:*	Gamma
Run Sequence:	1	*No. Isotopes:*	12
Batch Data Report:	LALLW0961	*Item Name:*	Empty TRU Drum 85/55-OP

Net Weight (kg):	2659.1		
Pu Mass (g):	1.79E-01	±	4.71E-02
Total Activity (Ci):	1.73E-02	±	4.11E-03
TRU Alpha Activity (Ci):	1.73E-02	±	4.11E-03
TRU Conc (nCi/g):	6.49E+00	±	1.54E+00
LLW Conc (nCi/g):	6.61E+00	±	1.54E+00
Pu239 Equivalent Activity (Ci):	1.73E-02	±	4.14E-03
Pu239 FGE (g):	1.69E-01	±	4.70E-02
Decay Heat (W):	5.45E-04	±	1.32E-04

Nuclide	Mass (g)	Activity (Ci)	Activity Uncertainty (Ci)	MDA (Ci)
Am241	1.20E-03	4.17E-03	2.76E-03	
Cm243	3.43E-08	1.79E-06	6.47E-07	
Cs137	4.60E-09	4.05E-07	9.85E-08	
Np237	9.57E-04	6.82E-07	1.80E-07	
Pu238	<LLD	<LLD	0.00E+00	3.11E-04
Pu239	1.69E-01	1.06E-02	2.96E-03	
Pu240	1.08E-02	2.48E-03	6.92E-04	
Pu241	<LLD	<LLD	0.00E+00	3.74E-02
Pu242	<LLD	<LLD	0.00E+00	1.43E-07
Sr90	2.93E-09	4.05E-07	9.85E-08	
U234	<LLD	<LLD	0.00E+00	2.27E-08
U235	<LLD	<LLD	0.00E+00	3.94E-10

Fig. 5. Example of a RDS for a composite virtual drum analysis

DEVELOPMENT TIME CONSIDERATIONS

While the screening and spectral summing approach once established can reduce the analyst resource demands and save a significant amount of analysis time, the time required developing

Radioassay Data Sheet

Assay Conclusion: LLW

MDAs Set with MT and Ref Isotope: PU52 (Pu239), U(NAT) (None)

Container ID:	90197	*Site ID:*	LANL
Revision No:	0	*Procedure:*	Q2/SNAP
File Name:	[illegible]	*Software 1:*	SnapV1.13
Detector:	Minnie	*Software 2:*	Peak Doctor v1.0
Assay Date and Time:	10/19/2011 0:00	*Assay Method:*	Gamma
Run Sequence:	1	*No. Isotopes:*	12
Batch Data Report:	LALLW0961	*Item Name:*	Empty TRU Drum 85/55-OP

Net Weight (kg):	66.5		
Pu Mass (g):	4.48E-03	±	7.45E-03
Total Activity (Ci):	4.31E-04	±	6.49E-04
TRU Alpha Activity (Ci):	4.31E-04	±	6.49E-04
TRU Conc (nCi/g):	6.49E+00	±	9.77E+00
LLW Conc (nCi/g):	6.61E+00	±	9.77E+00
Pu239 Equivalent Activity (Ci):	4.33E-04	±	6.55E-04
Pu239 FGE (g):	4.22E-03	±	7.43E-03
Decay Heat (W):	1.36E-05	±	2.09E-05

Nuclide	Mass (g)	Activity (Ci)	Activity Uncertainty (Ci)	MDA (Ci)
Am241	3.00E-05	1.04E-04	4.37E-04	
Cm243	8.57E-10	4.48E-08	1.02E-07	
Cs137	1.15E-10	1.01E-08	1.56E-08	
Np237	2.39E-05	1.71E-08	2.85E-08	
Pu238	<LLD	<LLD	0.00E+00	7.77E-06
Pu239	4.21E-03	2.65E-04	4.68E-04	
Pu240	2.70E-04	6.20E-05	1.09E-04	
Pu241	<LLD	<LLD	0.00E+00	9.34E-04
Pu242	<LLD	<LLD	0.00E+00	3.57E-09
Sr90	7.34E-11	1.01E-08	1.56E-08	
U234	<LLD	<LLD	0.00E+00	5.68E-10
U235	<LLD	<LLD	0.00E+00	9.84E-12

Fig. 6. Example of a RDS for an individual drum based on the spectral summing

the technical details and tools for the execution must be considered. Thresholds had to be calculated for the screening and two fairly elaborate spreadsheet utilities had to be developed for the overall application; one spreadsheet required formal V&V. Building the spreadsheet utilities and performing the associated V&V was the larger part of the development effort. This effort took several weeks overall before June 16, 2011 and benefited from utilizing a spectrum reader utility previously written for another project. Without the available spectrum reader, the

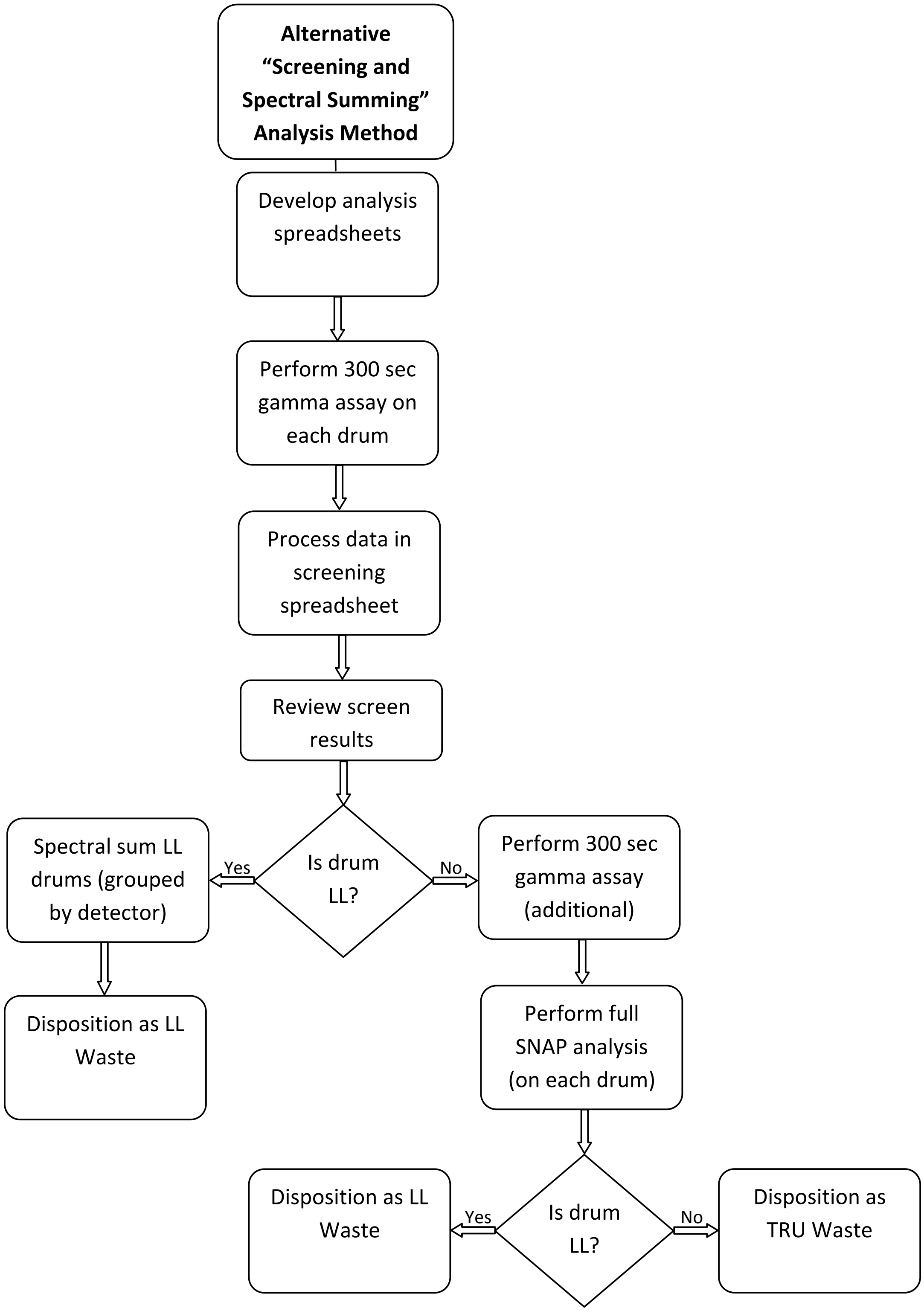

Fig. 7. Flowchart showing the main steps in the alternative screening spectral summing analysis methodology

development could have easily taken twice as long. Also, while overall analysis and review time was reduced, the daily quality assurance reviews of the screen results required more day to day attention than is required in the standard assay/analysis approach. This type of initial development time is probably only justified if a large number of similar items need to be measured.

CONCLUSION

The standard measurement and analysis methodology (using gamma spectroscopy) to achieve radiological characterization of waste drums at LANL is well established to rigorously meet the key requirement for LLW disposition. However, this approach can be time and resource intensive, especially when dealing with large populations. Instead of the standard approach, drum screening and spectral summing was applied to a large population of empty 55-gallon drums (often over-packed in 85-gallon drums) that formerly held TRU waste. Shorter 300 second screening measurements reduced the overall assay time for drums screened as LL, while final measurements for drums screened as TRU were suitable for the standard analysis method. Automated spreadsheets were developed (and required) to facilitate the screening of drums in real time and to spectral sum drums screened as LL that met specified requirements. Quality assurance measures were put in place to verify that the drum screening and spectral summing results were effective and met the key requirements for TRU/LL determination and LL disposition. Approximately 1000 drums were processed using the screening and spectral summing methodology over a 4 1/2 month span in 2011. Overall measurement and analysis time was reduced by applying the screening and spectral summing to the 2011 project. However, consideration must be given to the preparation time required for such operations (e.g., the development of automated spreadsheets or alternate analysis tools) when deciding if screening and spectral summing is the best approach for a particular project. The screening and spectral summing technique has proven to be a useful method available for meeting the radioactive characterization needs for large populations of sufficiently similar waste items at LANL and continues to be applied in ongoing operations.

REFERENCES

1. K.M. Gruetzmacher, et al., “Application of Spectral Summing to Suspect Low-Level Debris Drums at Los Alamos National Laboratory”, 51st Annual Meeting of the Institute of Nuclear Materials Management, Baltimore, MD, July 11 – July15, 2010.

2. K.M. Gruetzmacher, et al.,” Using Spectral Summing to Determine Radionuclide Activity for LANL Waste Drums Previously Containing Transuranic Waste”, 53rd Annual Meeting of the Institute of Nuclear Materials Management, Orlando, FL, July15 – July 19, 2012.

3. M.J. Clapham, J.V. Seamans and R.E. Arbon, “An NDA Technique for the Disposition of

Mixed Low Level Waste at the Advanced Mixed Waste Treatment Project", WM'06 Conference, Tucson, AZ, February 26 - March 2, 2006.

4. ENV-WGS-12-004, "MaestroSpecSum_.xltm Software Quality Assurance Documentation and Testing", LANL document, June 19, 2012.