Radioactive Waste Characterization Strategies; Statistical Comparison Dose to Curie and Gamma Spectroscopy Analysis Methods – 14229

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ABSTRACT

In the coming fiscal years of potentially declining budgets, Department of Energy facilities such as the Los Alamos National Laboratory (LANL) will be looking to reduce the cost of radioactive waste characterization, management, and disposal processes. At the core of this cost reduction process will be choosing the most cost effective, efficient, and accurate methods of radioactive waste characterization.

The majority of Generators at LANL have historically relied upon Acceptable Knowledge or Process Knowledge (AK/PK) to characterize their low level radioactive waste (LLRW) streams. As a sole tool for radiological waste characterization, this method of characterization has many drawbacks that result in inaccurate radiological waste stream characterization.

To supplement the LANL AK/PK analytical method, LANL has a well-developed and cost effect Non-Destructive Analysis (NDA) program. However, many small LANL Generators do not have the funding resources to utilize this program. In an effort to find alternate characterization methods to supplement the Generator AK/PK process, LANL has been working to implement the use of dose to curie software to increase the accuracy of the radiological waste stream characterization process.

This paper details the efforts to demonstrate the effectiveness and accuracy of the dose to curie characterization method by analyzing and comparing radiological modeling results to a source of known radiological content and also to the NDA results from gamma spectroscopy. This will show that dose to curie software programs can be used as a cost effective supplement to the AK/PK process and a certifiable alternative to Generators who do not have the resources to employ a NDA program.

INTRODUCTION

It is essential that laboratories within the Department of Energy complex implement a certifiable radioactive waste stream characterization program that takes into consideration implementation costs, ALARA and other hazards exposure, and accuracy in support of the core missions of the national weapons program and fundamental science and research. Reliable radiological characterization is the foundation of a waste management program allowing for the proper packaging, transportation, and disposal or radioactive waste at a minimal cost.

The purpose of this paper is to demonstrate that a reliable and cost effective dose to curie program is an effective and accurate method of radioactive waste analysis. A certified dose to curie analytical program can provide supplemental characterization data to AK/PK and significantly improve the quality of radiological waste characterization data while reducing overall characterization costs.

METHODS

Basic Radiological Waste Stream Characterization

Every radiological waste stream slated for disposal at a licensed facility must undergo radiological characterization. Each Generator must certify that the waste stream meets local, state, and Federal regulations for the packaging, storage, transportation, and disposal of the waste. This requires that waste streams must be characterized and certified to ensure compliance to Environmental Protection Agency (EPA) permits, Department of Energy (DOE) Orders, Nuclear Regulatory Commission (NRC) regulations, DOT regulations, and Treatment, Storage, and Disposal Facility (TSDF) Waste Acceptance Criteria (WAC) as applicable.

A Generator must weigh the benefits and drawbacks of the various analytical methods available to their program and determine which method or combination of methods will meet the regulatory requirements that the Generator is bound to in the most cost effective and safe manner possible. To reach this regulatory and safety threshold, the Generator must consider the cost benefit analysis (basic materials costs, overall program operations costs, man-hours per sample analyzed, etc.), radiation exposure As Low As Reasonably Achievable (ALARA) program considerations, Industrial Health and Safety risks, and overall analytical confidence level.

LANL Waste Generators rely heavily on AK/PK when determining the total radiological content of their waste streams. This holds true for Generators who maintain complete control of the waste generating process. These Generators know exactly what radiological isotopes and quantity they use in the process that generated the waste. Other Generators, such as those responsible for legacy waste cleanup projects or are owners of orphaned waste also rely extensively on AK/PK to characterizing their waste using limited sampling and historical data to quantify their waste stream’s radiological content.

Generator’s in the latter category run the risk of incorrectly characterizing the radiological constituents of their waste stream. A certified and validated dose to curie characterization process can minimize the risk of mischaracterizing a radiological waste stream.

The use of a dose to curie conversion software program can be a very effective method of characterizing a Generator’s Low Specific Activity (LSA) or Surface Contaminated Object (SCO) radiological waste stream and coupled with either AK/PK, gamma or neutron spectroscopy, or certified laboratory analytical. This method can in most circumstances be used to certify your waste stream for disposal depending on the TSDF WAC.

For example, a waste stream with a known history of radiological contamination, such as a drainage system that has had minimal radiological characterization (such as smears that have been analyzed for radiological isotopes) would lend itself to the use of a dose to curie conversion software program analysis. In this scenario, the material’s radiological scaling factors can be determined based on the smear analysis and can easily be applied to the waste once the waste has been removed and packaged.

Another example of a radiological waste stream that lends itself to the use the use of a dose to curie conversion software program is sealed sources that no longer have their initial manufacturing data and only the radioisotope is known. In this case, knowing the specific isotope, geometry, and mass for a single source and measuring the dose rate of that source will allow a Generator to use a dose to curie conversion software program with a high level of accuracy and allow a Generator to certify the waste stream for packaging, transportation, and disposal.

The advantages of using a dose to curie conversion software program are significant. Dose to curie data can provide an inexpensive method of radiological characterization with minimal radiological exposure and low industrial hygiene hazards exposure. Dose to curie methodology performs best as the primary method of analysis when coupled with solid AK/PK. Dose to curie methodologies excel in validating waste streams that come from a well-defined process where mass balance procedures and strict materials accountability can be demonstrated and for waste streams where some basic laboratory sampling has determined the isotopic ratios. Furthermore if the Generator combines the results of the dose to curie data with supplemental analytical such as high quality AK/PK, gamma spectroscopy, or laboratory analytical, the Generator can confidently certify the waste stream for disposal.

The disadvantages of using dose to curie data include the fact that some exposure to radiation is typically necessary to get detailed and accurate radiological survey maps for modeling. Dose to curie models tend to view materials that have been packaged as homogenous and without solid AK/PK, supplemental NDA analysis, or laboratory analytical, this method alone may not meet the certification requirements for TSDF disposition.

In addition, the accuracy of a dose to curie program can be greatly reduced by radiological hot spots located within the package, a heterogeneous waste material type, inaccurate modeling, and inappropriate selection of waste density matrices.

Therefore, based on the above analysis, LANL has determined that the most effective use of a dose to curie software tool as the primary method of analysis and characterization can be used best with waste streams that come from a well-defined process where mass balance procedures and strict materials accountability can be demonstrated and/or combined with supplemental gamma spectroscopy, or laboratory analytical   In cases where a large, uniform waste stream is being characterized, it may be sufficient to use a representative sample (e.g. 10% of the waste stream) versus sampling 100% of the waste containers.

LANL Waste Characterization Engineers and Subject Matter Experts developed the dose to curie methodology used in this paper by evaluating a known, certified source packaged into a 55 gallon drum in a well-defined matrix. The sources used in the drum model were calibration sources (**Table 1.0**) of Am241 (62.702 uCi) and Eu152 (39.369 uCi) arrayed in a spiral formation and embedded into a foam matrix to simulate a cellulose based waste stream (**Figure 1.0**).

Decay correct To:	11/5/2013									
		Activity (µCi)		Uncertainty (1σ)		Decay Corrected Activity			Uncertainty (99%)	
Source ID	Reference Date	^{241}Am	^{152}Eu	^{241}Am	^{152}Eu	^{241}Am	^{152}Eu		^{241}Am	^{152}Eu
63723	4/1/2005	5.756	5.682	1.35%	1.36%	5.677	3.634		3.47%	3.49%
63724	4/1/2005	5.284	5.794	1.35%	1.36%	5.212	3.705		3.48%	3.50%
63726	4/1/2005	5.523	5.581	1.35%	1.36%	5.447	3.569		3.48%	3.50%
63727	4/1/2005	5.606	5.408	1.35%	1.36%	5.529	3.459		3.47%	3.50%
63728	4/1/2005	5.854	6.094	1.34%	1.34%	5.774	3.897		3.46%	3.44%
63960	4/1/2005	5.915	5.599	1.36%	1.39%	5.834	3.581		3.50%	3.57%
46487	9/25/2003	4.955	4.910	1.35%	1.36%	4.875	2.902		3.48%	3.50%
46488	9/25/2003	4.935	4.842	1.35%	1.36%	4.856	2.862		3.48%	3.50%
46489	9/25/2003	4.944	4.895	1.35%	1.36%	4.865	2.893		3.48%	3.50%
46490	9/25/2003	4.916	4.985	1.35%	1.36%	4.837	2.946		3.48%	3.50%
46491	9/25/2003	4.986	5.025	1.35%	1.36%	4.906	2.970		3.48%	3.50%
46492	9/25/2003	4.970	4.994	1.35%	1.36%	4.890	2.952		3.48%	3.50%
	SUBTOTAL	63.644	63.809			62.702	39.369			

Table 1.0 Certified Calibration Sources in Model Matrix

This configuration allowed LANL to use a certified radiological curie value, a well-defined geometry of the standard 55 gallon drum, and a well-defined density of the materials in the drum as a known baseline to compare the dose to curie model against as well as compare the results against the LANL certified NDA analysis of the same drum. To be considered accurate enough to certify the waste for transportation and disposal of the radioactive materials, LANL Characterization Engineers established a +/- 50% (of the certified source values) threshold for evaluating the dose to curie analytical results.

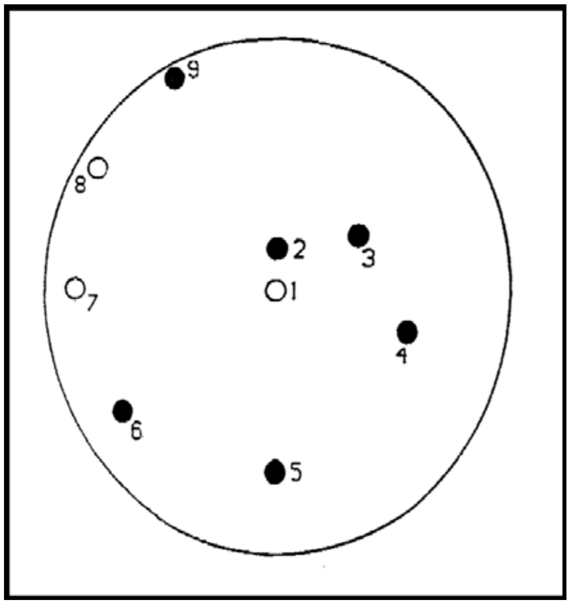


Figure 1.0 Top View, Cross Section of Calibration Drum

Basic Waste Stream Analysis and Characterization using Dose to Curie Methods

To accurately construct the drum model in the LANL selected dose to curie software program, Characterization Engineers had to perform several field activities in order to enter the required data into the selected dose to curie program.

The Characterization Engineers first evaluated the radiological content of the certified mixed gamma standard source. The source drum selected contained (62.702 uCi) of AM241 and (39.369 uCi) of Eu152 decay corrected to November 5, 2013. The sources were arranged in a solid foam matrix of (0.9) gr/cm3 to simulate a cellulose waste drum being characterized for disposal that can be found in **Figure 1.0** with associated activities listed in **Table 1.0**.

LANL personnel then carefully constructed the geometry of the standard 55 gallon drum in the dose to curie program. The geometry construction was based on the construction diagrams of a standard 55 gallon drum as seen in **Figure 2.0**.

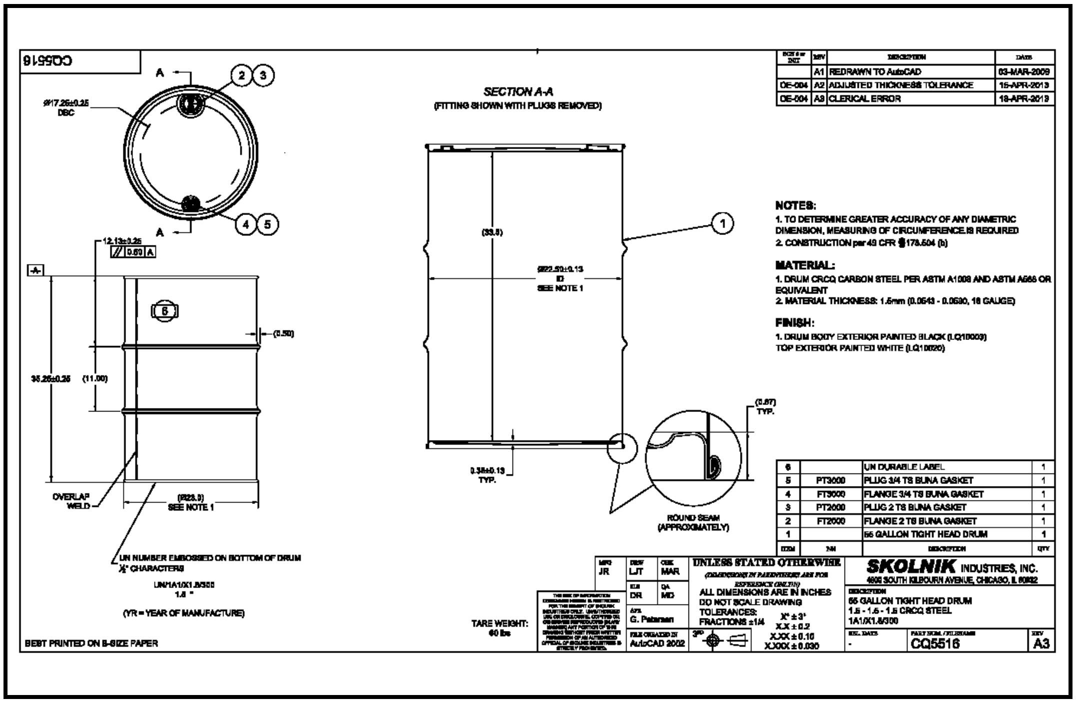


Figure 2.0 Construction Drawing Standard 55 Gallon Drum

LANL personnel then took multiple dose readings to comprehensively characterize the radiation does coming from the selected drum. These dose readings allowed the Characterization Engineers to construct a three dimensional dose model that showed areas of elevated dose readings (hot spots) and the overall uniform distribution of the radiological isotopes in the drum matrix. The drum was then divided into four quadrants and each quadrant was analyzed as four individual drums. These dose readings can be found in **Table 2.0**.

One Meter Model

CID	Survey Point	1 M(uR/hr)	1 M Average (uR/hr)	Minus Background
EU-001	1M-01	28	28.25	10.25
	1M-02	29		
	1M-03	28		
	1M-04	28		

One Foot Model

CID	Survey Point	1 FT (uR/hr)	1 FT Average (uR/hr)	Minus Background
EU-001	1F-01	170	170	152

On Contact Q1 Model

CID	Survey Point	1 OC (uR/hr)	1 OC Average (uR/hr)	Minus Background
EU-001	1T-01	213	221	203
	1M-01	246		
	1B-01	204		

On Contact Q2 Model

CID	Survey Point	1 OC (uR/hr)	1 OC Average (uR/hr)	Minus Background
EU-001	2T-01	179	222.00	204
	2M-01	207		
	2B-01	280		

On Contact Q3 Model

CID	Survey Point	1 OC (uR/hr)	1 OC Average (uR/hr)	Minus Background
EU-001	3T-01	232	274.67	256.67
	3M-01	290		
	3B-01	302		

On Contact Q4 Model

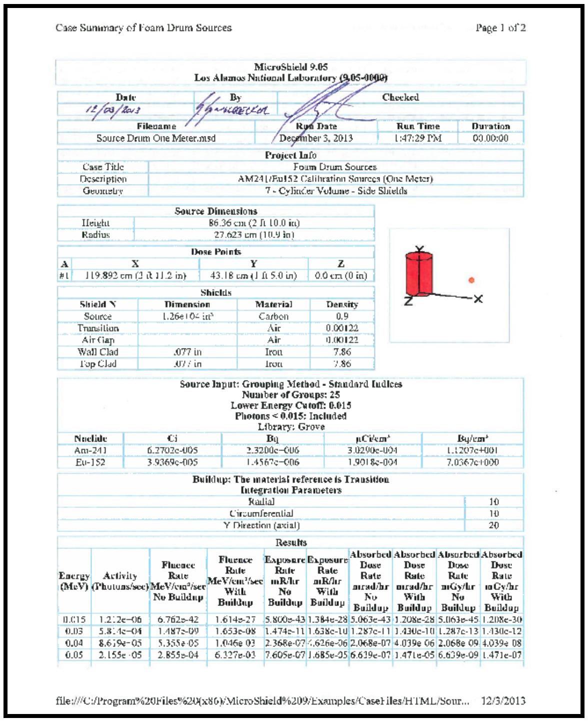
CID	Survey Point	1 OC (uR/hr)	1 OC Average (uR/hr)	Minus Background
EU-001	4T-01	242	246.67	228.67
	4M-01	275		
	4B-01	223		

Highest On Contact Q4

CID	Survey Point	1 OC (uR/hr)	1 OC Average (uR/hr)	Minus Background
EU-001	HT-01	330	313.33	295.33
	HM-01	300		
	HB-01	310		

Table 2.0 Dose Construction Mode

An example of the resulting dose to curie models can be found in **Figure 3.0** and indicates that the average curie content of the drum is (81.3 uCi) of Am241 and (51.1 uCi) of Eu152 as indicated in **Table 4.0**. This compares favorably with the known value of the source drum and also to the LANL certified NDA gamma spectroscopy analysis performed on this same drum. A full comparison of the analytical results can be found in **Table 4.0**.



Case Summary of Foam Drum Sources — Page 1 of 2

MicroShield 9.05 Los Alamos National Laboratory (9.05-0000)		
Date	By	Checked
12/03/2013		

Filename	Run Date	Run Time	Duration
Source Drum One Meter.msd	December 3, 2013	1:47:29 PM	00:00:00

Project Info	
Case Title	Foam Drum Sources
Description	AM241/Eu152 Calibration Sources (One Meter)
Geometry	7 - Cylinder Volume - Side Shields

Source Dimensions	
Height	86.36 cm (2 ft 10.0 in)
Radius	27.623 cm (10.9 in)

Dose Points			
A	X	Y	Z
#1	119.892 cm (3 ft 11.2 in)	43.18 cm (1 ft 5.0 in)	0.0 cm (0 in)

Shields			
Shield N	Dimension	Material	Density
Source	1.26e+04 in³	Carbon	0.9
Transition		Air	0.00122
Air Gap		Air	0.00122
Wall Clad	.077 in	Iron	7.86
Top Clad	.077 in	Iron	7.86

Source Input: Grouping Method - Standard Indices
Number of Groups: 25
Lower Energy Cutoff: 0.015
Photons < 0.015: Included
Library: Grove

Nuclide	Ci	Bq	μCi/cm³	Bq/cm³
Am-241	6.2702e-005	2.3200e-006	3.0290e-004	1.1207e+001
Eu-152	3.9369e-005	1.4567e-006	1.9018e-004	7.0367e+000

Buildup: The material reference is Transition Integration Parameters	
Radial	10
Circumferential	10
Y Direction (axial)	20

Results

Energy (MeV)	Activity (Photons/sec)	Fluence Rate MeV/cm²/sec No Buildup	Fluence Rate MeV/cm²/sec With Buildup	Exposure Rate mR/hr No Buildup	Exposure Rate mR/hr With Buildup	Absorbed Dose Rate mrad/hr No Buildup	Absorbed Dose Rate mrad/hr With Buildup	Absorbed Dose Rate mGy/hr No Buildup	Absorbed Dose Rate mGy/hr With Buildup
0.015	1.212e-06	6.762e-42	1.614e-27	5.800e-43	1.384e-28	5.063e-43	1.208e-28	5.063e-45	1.208e-30
0.03	5.814e-04	1.487e-09	1.653e-08	1.474e-11	1.638e-10	1.287e-11	1.430e-10	1.287e-13	1.430e-12
0.04	8.619e-05	5.355e-05	1.046e-03	2.368e-07	4.626e-06	2.068e-07	4.039e-06	2.068e-09	4.039e-08
0.05	2.155e-05	2.855e-04	6.327e-03	7.605e-07	1.685e-05	6.639e-07	1.471e-05	6.639e-09	1.471e-07

file:///C:/Program%20Files%20(x86)/MicroShield%209/Examples/CaseFiles/HTML/Sour... 12/3/2013

Figure 3.0 Example of Completed Dose to Curie Model

DISCUSSION

The use of a dose to curie conversion software program can be a very effective method of characterizing a Generator’s Low Specific Activity (LSA) or Surface Contaminated Object (SCO) radiological waste stream and coupled with either AK/PK, gamma or neutron spectroscopy, or certified laboratory analytical. This method can in most circumstances be used to certify your waste stream for disposal depending on the TSDF WAC.

The comparison of our dose to curie model described above compares favorably to the known source drum as well as analytical provided on the same drum by the LANL NDA gamma spectroscopy team.

This effectively demonstrates that a Generator, with minimum AK/PK and knowing the specific isotope, geometry, and mass for a single source and measuring the dose rate of that source will allow a Generator to use a dose to curie conversion software program with a high level of accuracy and allow a Generator to certify the waste stream for packaging, transportation, and disposal.

The dose to curie analytical method is also relatively inexpensive and cost effective. Like the

AK/PK method, the dose to curie method of analysis can be used cost effectively for small waste streams with only a few containers to be characterized and can also be used for large waste streams with thousands of containers with little impact on the overall cost of characterization analysis. However, the dose to curie method also relies heavily on sound AK/PK or supplemental analytical methods and alone cannot determine RCRA or TSCA constituents.

		Certified Ci		Calculated Ci		Isotopic Variance	
Model Drum #	Distance	Am241	Eu152	Am241	Eu152	Am241	Eu152
1M	One Meter	6.27E-05	3.94E-05	5.98E-05	3.76E-05	95.4%	95.4%
1F	One Foot	6.27E-05	3.94E-05	7.73E-05	4.86E-05	123.3%	123.4%
Q1	On Contact	6.27E-05	3.94E-05	7.98E-05	5.01E-05	127.3%	127.2%
Q2	On Contact	6.27E-05	3.94E-05	8.01E-05	5.03E-05	127.8%	127.7%
Q3	On Contact	6.27E-05	3.94E-05	1.01E-04	6.33E-05	161.1%	160.7%
Q4	On Contact	6.27E-05	3.94E-05	8.98E-05	5.64E-05	143.2%	143.1%

Table 3.0 Dose to Curie Radiological Data

CONCLUSIONS

The comparison of our dose to curie model described above compares favorably to the known source drum as well as analytical provided on the same drum by the LANL NDA gamma spectroscopy team.

However, the significant disadvantages of using dose to curie data include the fact that some exposure to radiation is typically necessary to get detailed and accurate radiological survey maps for modeling. Dose to curie models tend to view materials that have been packaged as homogenous and without solid AK/PK, supplemental NDA analysis, or laboratory analytical, this method alone may not meet the certification requirements for TSDF disposition.

In addition, the accuracy of a dose to curie program can be greatly reduced by radiological hot spots located within the package, a heterogeneous waste material type, inaccurate modeling, and inappropriate selection of waste density matrices. To mitigate these drawbacks, the LANL dose to curie programs will only be allowed for use by certified Waste Characterization Engineers, Subject Matter Experts, and others who are trained and qualified on the use and weaknesses of the software program.

This effectively demonstrates that a Generator, with minimum AK/PK and knowing the specific isotope, geometry, and mass for a single source and measuring the dose rate of that source will allow a Generator to use a dose to curie conversion software program with a high level of accuracy and allow a Generator to certify the waste stream for packaging, transportation, and disposal.

		Certified Ci		Calculated Ci		Isotopic Variance	
Model Drum #	Distance	Am241	Eu152	Am241	Eu152	Am241	Eu152
Composite Analysis	Composite	6.27E-05	3.94E-05	8.13E-05	5.11E-05	130%	130%
NDA Gamms Spec	On Contact	6.27E-05	3.94E-05	9.59E-05	5.47E-05	153%	139%

Table 4.0 Radiological Data Comparison to Gamma Spectroscopy Values

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