Radioactive Waste Characterization Strategies; Comparisons Between AK/PK, Dose to Curie Modeling, Gamma Spectroscopy, and Laboratory Analysis Methods- 12194

Steven J. Singledecker, Scotty W. Jones, Alison M. Dorries, George Henckel, Kathleen M. Gruetzmacher

Los Alamos National Laboratory, Los Alamos, New Mexico 87545

LA-UR 11-06923

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.

Central to every radioactive waste management program is an effective and accurate waste characterization program. Choosing between methods can determine what is classified as low level radioactive waste (LLRW), transuranic waste (TRU), waste that can be disposed of under an Authorized Release Limit (ARL), industrial waste, and waste that can be disposed of in municipal landfills. The cost benefits of an accurate radioactive waste characterization program cannot be overstated.

In addition, inaccurate radioactive waste characterization of radioactive waste can result in the incorrect classification of radioactive waste leading to higher disposal costs, Department of Transportation (DOT) violations, Notice of Violations (NOVs) from Federal and State regulatory agencies, waste rejection from disposal facilities, loss of operational capabilities, and loss of disposal options. Any one of these events could result in the program that mischaracterized the waste losing its ability to perform it primary operational mission.

Generators that produce radioactive waste have four characterization strategies at their disposal:

- Acceptable Knowledge/Process Knowledge (AK/PK)
- Indirect characterization using a software application or other dose to curie methodologies
- Non-Destructive Analysis (NDA) tools such as gamma spectroscopy
- Direct sampling (e.g. grab samples or Surface Contaminated Object smears) and laboratory analytical

Each method has specific advantages and disadvantages. This paper will evaluate each method detailing those advantages and disadvantages including;

- 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
- Overall Analytical Confidence Level

The concepts in this paper apply to any organization with significant radioactive waste characterization and management activities working to within budget constraints and seeking to optimize their waste characterization strategies while reducing analytical costs.

INTRODUCTION

It is essential that laboratories within the Department of Energy complex implement a cost effective radioactive waste characterization program that takes into consideration costs, ALARA and other hazards exposure, and accuracy in support of the core missions of the national weapons program and fundamental science and research.

The purpose of this paper is to compare the four methods of analysis listed above and provide an overall evaluation of each methods advantages and disadvantages. To evaluate cost, radiation exposure, and industrial hazards, a typical LLRW waste stream was developed and LANL resource costs were used as a baseline.

METHOD AND RESULTS

Basic Waste Stream Characterization for Disposal

Every waste stream slated for disposal at a licensed facility must undergo chemical and 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.

Basic Waste Stream Analysis and Characterization

To characterize a waste stream for radiological, Toxic Substance Control Act (TSCA), and Resource Conservation and Recovery Act (RCRA) constituents and create a Waste Profile Record for disposal, a Generator needs to undertake the following minimum waste evaluations:

- Review site or process history;
- Determine RCRA/TSCA constituents and concentrations;
- Perform isotopic evaluation including:
 - o determining isotopes of concern and isotopic concentration; and
 - comparing present isotopes to the isotopic limits of acceptance at the selected TSDF.
- Define the waste physical form;
- Evaluate DOT considerations; and
- Define TSDF WAC analytical requirements.

All of these items must be evaluated when creating a Waste profile record and choosing a disposal path.

Waste Stream Description for Baseline Comparison

The waste stream used to compare cost and potential exposure consists of fifty (50) B25 IP-II certified containers containing soil contaminated with radiological isotopes and RCRA/TSCA hazardous constituents. Each container reads approximately 5 mR/hr on contact.

Each analytical method was compared to the Environmental Protection Agency (EPA) Test Methods SW-846 to achieve a 95% confidence level in collected data.

Cost modeling is based on the most recent LANL Project Work Package, WBS# X1.06.14.02.04.03.

AK/PK Analytical Method Evaluation

Acceptable Knowledge/Process knowledge (AK/PK) consists of reviewing the known history of a waste site or waste generation process and defining the hazardous constituents based on this review. Using AK/PK to characterize a Generator waste stream can minimize the Generator's cost of characterization but is limited in what waste streams it can be used to certify.

For example, in medical and biological research requiring the use of radioisotopes and hazardous chemicals (such as H3 and C14 as tracer isotopes and toluene as a solvent), the researcher can define the precise amount of radiological materials needed and used in the experiment. When the experiment is complete, the researcher can certify the amount of isotopic materials and solvents used during the experiment and are now in the waste based on the use of mass balance and can certify the waste with high confidence for packaging, transportation, and disposal.

Another example of the mass balance method to support AK/PK is certified sources being prepared for disposal. If the source certification is available and/or the markings on the source are present, this type of information can be used to certify the isotopic content (after performing activity decay calculations) and concentration for transportation and disposal. Therefore AK/PK in these limited scenarios can be a very effective and accurate means of characterizing a Generator's waste.

The advantages of using AK/PK combined with a system of mass balance and strict inventory control are clear. AK/PK is relatively inexpensive usually requiring only the labor of a certified characterization professional to characterize the waste stream. There is no exposure to radiological or industrial hazards and no exposure to RCRA or TSCA constituents.

The disadvantages of using AK/PK as the sole method of characterization are numerous. For waste streams that are not well defined and where no systems of mass balance or inventory control were used, the waste stream characteristics cannot be certified and will most likely not meet the certification requirements for TSDF disposition or allow for adequate DOT characterization. [1]

Therefore, based on the above analysis, the most effective use of AK/PK 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. In addition, AK/PK will almost always play some baseline role in helping the Generator decide which final analytical method will be chosen to characterize the waste stream.

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. However, since the costs of employing the AK/PK method are relatively inexpensive, the Generator may decide to employ this method for all waste containers. A baseline cost evaluation for using AK/PK can be found in Table I.

Acceptable Knowledge/Process Knowledge Method Cost Analysis [2]										
	Cost per	Man-			Cost per	Radiological	IH			
Position	Hour	Hours	Tota	Base Cost	Sample	Exposure	Exposure	Accuracy		
								Mass Balance:		
Characterization Engineer	\$ 129.00	40	\$	5,160.00	NA	None/Low	None/Low	High		
Quality Assurance								Legacy Waste:		
Engineer	\$ 145.00	8	\$	1,160.00				Low		
		Total	\$	6,320.00						

Table I. AK/PK Analytical Method Cost Evaluation

Dose to Curie Analytical Method Evaluation

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.

However, in addition to the basic waste stream characterization requirements, a Generator must also consider the following additional requirements that will be required for using the dose to curie software for waste stream characterization:

- The Generator must determine the ratio of radiological isotopes in the waste stream. This will require some level of AK/PK, NDA, or direct, physical sampling;
- Health Physics personnel must perform radiological package surveys to develop an accurate dose rate model for the dose to curie software data input;
- The Generator must evaluate health and safety risks from radiological exposure and industrial hazards (i.e. pinch/crush potential) from the physical movement the packages;
- Have qualified personnel to accurately build the dose to curie model; and
- Have a qualified Radiological Engineer to validate the model and the results.

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 (an estimated radiation exposure analysis can be found in Table III). Dose to curie models tend to view materials that have been packaged as homogenous [3] and without solid AK/PK, supplemental NDA analysis, or laboratory analytical, this method alone may not meet the certification requirements for TSDF disposition. Furthermore, dose to curie data does not allow

for RCRA or TSCA analysis and certification without strict AK/PK or certified laboratory analysis and therefore may not allow for adequate DOT characterization.

Therefore, based on the above analysis, 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. However, in small and large waste streams, since the costs of employing the dose to curie method are relatively inexpensive, the Generator may decide to employ this method for all waste containers. A baseline cost evaluation for using the dose to curie analytical method can be found in Table II.

Dose to Curie Software Method Cost Analysis [2]										
			Man-					Radiological	IH	
Position	Cost p	er Hour	Hours	Total	Base Cost	Cost	oer Sample	Exposure	Exposure	Accuracy
										Mass
								Medium-		Balance:
Characterization Engineer	\$	129.00	40	\$	5,160.00	\$	586.40	High	None	High
										Legacy
										Waste:
Quality Assurance Engineer	\$	145.00	8	\$	1,160.00					Med-High
Dose to Curie Software										Analytical
Data Technician	\$	97.00	100	\$	9700.00					Data: High
Radiological Engineer	\$	165.00	20	\$	3,300.00					
Radiological Controls										
Technician	\$	125.00	50	\$	6,250.00					
Waste Management										
Technician	\$	75.00	50	\$	3,750.00					
			Total	\$	29,320.00					

Table II. Dose to Curie Analytical Method Cost Evaluation

Dose to Curie Software Method Exposure Analysis [4]									
Position	Dose Rate @ 1 Ft (mRad/hr)	Man- Hours	Estimated Exposure (mRem)						
Radiological Controls Technician	5	50		250					
Waste Management Technician	5	20		100					
		Total		350					

Table III. Dose to Curie ALARA Evaluation

Gamma or Neutron Spectroscopy Non-Destructive Analysis Analytical Method Evaluation

Non-Destructive Analysis (NDA) using gamma or neutron spectroscopy analysis can be a very effective method of characterizing a Generator's waste stream where little information is known about the radiological content and can in most radiological waste stream circumstances, be used to certify the Generator's waste stream for disposal depending on the TSDF WAC.

For example, gamma spectroscopy can be used to determine what isotopes are present within a legacy or orphan waste stream with gamma emitting radiological constituents.

Gamma or neutron spectroscopy can also be used with a high level of confidence on a waste stream with a known history of TRU radiological contamination where the isotopic distribution is well-defined, such as plutonium research and manufacturing. In this scenario, the radiological content and concentration can be determined based on the NDA results. [5]

Another example of a radiological waste stream that lends itself to the use of gamma spectroscopy is sealed sources that no longer have their initial manufacturing data and the radioisotope is unknown. In this case, gamma spectroscopy can define the radioisotope and total radiological content with a high level of accuracy and allow a Generator to certify the waste stream for packaging, transportation, and disposal.

However, in addition to the basic waste stream characterization requirements, a Generator must also consider the following additional requirements that will be required for using NDA for waste stream characterization:

- The Generator must have access to or support a certified NDA program and gamma spectroscopy equipment;
- The Generator must evaluate health and safety risks from radiological exposure and industrial hazards (i.e. pinch/crush potential, gamma spectroscopy equipment set up) from the physical movement the packages and performance of the NDA analysis;
- Have qualified personnel to accurately build the NDA model; and
- Have a qualified Radiological Engineer to validate the model and the results.

There are specific advantages of using the NDA tool for waste certification. NDA can provide high quality and accurate isotopic data with minimal exposure for waste streams known to have only isotopic constituents and, depending on the TSDF WAC, can be used to certify the waste for transportation and disposal. [6]

The disadvantages of using the NDA tool include the fact that some exposure to radiation is typically necessary when performing the gamma spectroscopy analysis (an estimated radiation exposure analysis can be found in Table V). NDA will not provide any RCRA/TSCA data on the waste stream or identify low level beta-gamma emitters or alpha emitters requiring the Generator to either rely on AK/PK or physical sampling and analysis to rule out or confirm these constituents. For waste streams that are suspected to contain both radiological and hazardous constituents, NDA alone may not meet the certification requirements for TSDF disposition.

Therefore, based on the above analysis, the most effective use of the NDA tool as the primary method of analysis and characterization can be used best with waste streams known to solely contain radiological constituents. In cases where a 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. A baseline cost evaluation for using NDA can be found in Table IV.

NDA (Gamma Spectroscopy) Method Cost Analysis [2]										
			Man-					Radiological	IH	
Position	Cost pe	r Hour	Hours	Tota	al Base Cost	Cost pe	er Sample ⁽¹⁾	Exposure	Exposure	Accuracy
										Mass
										Balance:
Characterization Engineer	\$	129.00	24	\$	3,096.00	\$	782.60	Medium	None	High
										Legacy
										Waste:
Quality Assurance Engineer	\$	145.00	8	\$	1,160.00					High
NDA Gamma Spec Software	\$									Analytical
Data Technician	97.00		17	\$	1,649.00					Data: High
NDA Gamma Spec Operator	\$	165.00	100	\$	16,500.00					
Radiological Controls										
Technician	\$	125.00	25	\$	3,125.00					
Waste Management	\$									
Technician	75.00		100	\$	7,500.00					
Programmatic Equipment	\$									
Costs ⁽²⁾	61.00		100	\$	6,100.00					
		•	Total	\$	39,130.00					

Note (1): LANL average cost per sample: \$366.54/sample

Note (2): LANL M&E and Contracts Annual Costs: \$250,000

Table IV. NDA Analytical Method Cost Evaluation

NDA (Gamma Spectroscopy) Method Exposure Analysis [4]									
Position	Dose Rate @ 1 Ft (mRad/hr)	Man- Hours	Estimated Exposure (mRem)						
Radiological Controls Technician	5	6	30						
Waste Management Technician	5	17	85						
NDA Gamma Spec Operator	5	8	40						
Characterization Engineer	5	0.5	2.5						
		Total	157.5						

Table V. NDA ALARA Evaluation

Physical Samples with Certified Laboratory Analytical Method Evaluation

Physical samples taken from the waste stream and analyzed by a certified laboratory can provide the most detailed and defensible data for characterizing and certifying a Generator's waste stream. Detailed laboratory analysis will define what radionuclides are present as well as isotopic concentration. Furthermore, the laboratory analysis of physical samples will define what hazardous constituents are present in the waste. This data can be used to certify the Generator's waste stream for storage, transportation, and disposal.

The advantages of using certified laboratory analysis for a waste stream include defensible analytical data, clearly defined radiological and hazardous constituents.

In addition, the data received for the waste will meet the certification requirements for TSDF disposition and allow for adequate DOT characterization.

The disadvantages of using certified laboratory analysis include the fact that exposure to radiation and potential hazardous components is necessary to pull the physical samples (an estimated radiation exposure analysis can be found in Table VII). Furthermore, unless the waste stream can be shown to be homogenous, it may be difficult or impossible to obtain a representative physical sample for analysis. In addition, costs tend to be much higher than the other methods discussed in this paper.

Therefore, based on the above analysis, the most effective use of laboratory analytical as the primary method of analysis and characterization can be used best with legacy waste streams with little or no AK/PK. 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. A baseline cost evaluation for using laboratory analytical can be found in Table VI.

		Ph	sical San	nples with Laborator	y Analytical Meth	od Cost Analysis	[2]		
			Man -	•	,	,			
	Cost	t per Unit	Hour			Cost per	Radiological	IH	
Position	Rate	9	S	Total Base Cost		Sample (1)	Exposure	Exposure	Accuracy
Characterization Engineer	\$	129.00	24	\$	3,096.00	\$ 3,434.42	High	High	Mass Balance: High
Quality Assurance Engineer	\$	145.00	8	\$	1,160.00			Ü	Legacy Waste: High
Sample and Analysis Program Manager	\$	97.00	17	\$	1,649.00				Analytical Data: High
Integrated Word Document Planner	\$	165.00	100	\$	16,500.00				
Sample Control Technicians	\$	85.00	80	\$	6,800.00				
Sample Management Office Support	\$	77.00	8	\$	616.00				
Radiological Controls Technician	\$	125.00	40	\$	5,000.00				
Industrial Hygiene Technician	\$	110.00	40	\$	4,400.00				
Waste Management Technician	\$	75.00	100	\$	7,500.00				
Sample Costs	\$ 2	2,500.00	50	\$	125,000.00				
			Total	\$	171,721.00				

Table VI. Certified Laboratory Analytical Cost Evaluation

Physical Samples with Laboratory Analytical Method Cost Analysis [4]								
Position	Dose Rate @ 1 Ft (mRad/hr)	Man- Hours	Estimated Exposure (mRem)					
Radiological Controls Technician	5	10	50					
Waste Management Technician	5	20	100					
Sample Control Technicians	5	100	500					
Industrial Hygiene Technician	5	10	50					
		Total	700					

Table VII. Laboratory Analytical ALARA Evaluation

DISCUSSION

Each of the analytical methods discussed above come with clear advantages and disadvantages. AK/PK is an inexpensive method of characterization and can be used cost effectively for small waste streams with only a few containers to be characterized and large waste streams with thousands of containers with little impact on the overall cost of characterization analysis. This makes the AK/PK method cost effective and flexible particularly with waste streams where the waste generation process is clearly define and the hazardous materials introduced into the waste were clearly controlled and accounted for. However, AK/PK is inadequate for legacy or orphaned waste where no knowledge of the waste stream exists to perform an AK/PK analysis or where the hazardous materials introduced to the waste stream were not controlled and/or accounted for.

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.

Gamma or neutron spectroscopy NDA can generate high quality radiological data and for waste streams with no known RCRA or TSCA constituents, can be used to certify radiological waste streams for packaging, transport and disposal. However, NDA alone cannot determine RCRA or TSCA constituents where AK/PK may indicate that they are present. Furthermore, for large waste streams with many containers, the costs of implementing a NDA characterization program may prove to be beyond the budget of the project. Based on this cost limitation, the Generator my decide to perform representative sampling of the waste population instead of sampling all of the containers leading to a potential reduction of analytical confidence level.

Physicals samples analyzed by a certified laboratory can provide the highest level of analytical confidence to the Generator particularly if the waste stream can be shown to be homogenous and well defined. However, pulling physical samples tends to be expensive and expose personnel to radiological and industrial hazards. Furthermore, with large, heterogeneous waste streams, 100% sampling of all waste containers will not guarantee accurate analytical and the cost of sampling every individual containers in such waste streams are prohibitively expensive.

The answer to which analytical method to choose will vary based on the type and volume of the waste stream a Generator is dealing. Waste streams will vary significantly with waste types ranging from highly controlled and well defined waste streams from a production line to legacy or orphan waste streams with little or no data available for analysis. By carefully evaluating their waste stream and combining aspects of the various methods detailed above to minimize costs and personnel exposure while maximizing their overall analytical confidence level and confidence, the Generator can effectively tailor their sampling methodology to meet the TSDF WAC will controlling and minimizing their analytical costs.

REFERENCES

- 1. Hazardous materials Regulations, 49 Code of Federal Regulations, Parts 105-180, July 15, 2011.
- 2. Steven J. Singledecker. Jackie H. Hurtle, and Andrew M. Montoya, Los Alamos National Laboratory FY2012 Project Work Package, August 18, 2011.
- 3. Grove Software, Incorporated. MicroShield User's Manual, January 2011.
- 4. Shleien, Bernard. The Health Physics and Radiological Health Handbook, Revised Edition. Scinta, Inc., 1992.
- 5. Sher, R. and Untermeyer II S. The Detection of Fissionable Materials by Nondestructive Means. The American Nuclear Society, 1980.
- 6. Gilmore, Gordon. Practical Guide to Gamma-Ray Spectroscopy, 2nd Edition. John Wiley and Sons LTS, 2008.