Recounting History through Radioassay

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ABSTRACT

This paper describes a proposed method for using historical documentation to identify unknown wastes resulting from retrieving suspect transuranic (TRU) waste. Identification is accomplished by a historical review of radionuclides identified by radioassay, along with the project controls used to ensure an accurate segregation of TRU from low-level waste (LLW). This paper presents an historical perspective on the identification of radionuclides at the Hanford Site from various waste generators of suspect TRU waste with an emphasis on the Data Quality Objectives (DQO's) and project controls used to ensure the waste is properly classified as TRU or LLW.

DISCUSSION

Introduction

A key component of Hanford Site cleanup is the Waste Retrieval Project (WRP) within the Waste Stabilization and Disposition Project. The scope of the WRP is to remove or retrieve, classify, and characterize suspect TRU solid wastes buried between 1970 and 1987 on the Hanford Site. Suspect TRU waste was received from many different on-site and off-site waste generators. Documentation on each waste generator is compiled into an Acceptable Knowledge (AK) data package. Radioassay of up to 5,000 retrieved waste containers (primarily 210 liter (55-gallon) drums) per year is the critical step in classification of the waste as TRU or LLW. History of the Hanford Site and various nuclear programs may be tracked through the identification of radionuclides in the suspect TRU waste. Initially, most retrieved waste

containers were in good condition. However, for badly corroded containers, or containers with illegible markings, or containers with no labels, radioassay provides a useful tool to attempt to associate the waste container with the correct AK documentation to identify the waste generator. Wastes are considered unknown when they cannot be associated with the burial records or AK documentation. A few of the varied waste processes and radionuclide mixtures are described below:

- Weapons, fuel, and research-grade plutonium containing up to 23 percent Pu-240.
- Curium and plutonium-238 heat source material.
- Plutonium-uranium mixed oxide (MOX). This waste stream consists of fuel rod cladding and cans, and MOX fuel in the form of pellets, powder, and scrap (waste from the production of MOX). There is significantly more uranium than plutonium.
- Waste associated with the recovery of special nuclear materials including americium, curium, and californium.
- Waste from research and development of plutonium-bearing fuels including metallurgical examination and chemical separation of materials.
- Actinide separation studies including thorium and neptunium separations.
- Specific isotope isolation such as curium and promethium isotopes.
- Thorium-232 uranium-233 fuel cycle.

Suspect TRU Wastes were placed in burial trenches from 1970 through 1987 for future retrieval. Fig. 1 shows a typical configuration of a burial trench containing 210 liter drums of suspect TRU wastes. Other designs and configurations including other waste containers have been used to store the suspect TRU wastes. In the more recent waste configuration identified in Fig. 1, suspect wastes were tracked by module charts with a module consisting of a 12 by 12 drum array up to 4 drums high. The position of a drum within the module can be tracked by the length (position 1 through 12), width (position A through L), and height (Tier 1 through 4). For example, a drum identified in position 12, position A, tier 4 would indicate the uppermost right corner of the module in Fig. 1. In older situations, the drum might be identified to a particular module or set of geographic coordinates without identifying a specific position in the module. This system of module charts provides an effective tool in associating a retrieved waste container with the correct burial record and in resolving unknown containers when a drum is removed without legible labels or markings.

Fig 1. Typical TRU module in the 218-W-4C burial ground

The primary DQO or project goal is to ensure TRU wastes are never misclassified as LLW. The project employs a number of quality checks and project tools to verify and validate the accuracy of waste classification data. Traditional quality control checks such as control charting radioassay parameters are performed to ensure the assay system is operating properly. In addition, periodic independent assessments, surveillances, reviews, and assay of test standards are performed to validate and verify system operation. Radioassay measurements are performed in the field with mobile equipment to sort TRU from LLW. Based on field assay results, suspect TRU waste is separated into two pathways. TRU waste containers receive a certified radioassay to support disposal of the waste at the Waste Isolation Pilot Plant (WIPP) in New Mexico. The non-TRU waste stream is treated and disposed of on-site, according to regulatory requirements. As an example, one mixed LLW stream is super-compacted and macroencapsulated (grouted) in a waste box for disposal on-site. Approximately 47-percent of the retrieved suspect TRU waste is classified by radioassay as LLW.

Project Approach and Controls

Project management techniques are employed to ensure effective and efficient management of scarce resources. Project scope management ensures that the project includes all the work required, and only the work required to complete the project successfully.[1] Project Scope, and

specifically the scope of activities necessary to support radioassay of suspect TRU waste, is narrowly defined to accomplish project goals and cleanup of the Hanford Site.

Effective cost management includes resource planning, cost estimating, budgeting, and cost control. Tasks are defined, sequenced and scheduled to coordinate activities among the competing project goals. Project costs are stringently controlled to minimize impact to the project. Field radioassay screening is considerably less expensive than a certified radioassay for disposal of waste at the WIPP. Radioassay activities are currently limited to support removal of suspect TRU wastes from the 218-W-4C Burial Ground. A field assay is performed on 210 liter drums with records indicating the waste contains less than 1 gram plutonium and waste boxes with records indicating the waste contains less than 175 nCi/g TRU. Those containers with records indicating the waste contains greater than these quantities of plutonium, and those containers determined to be TRU waste by field assay, are sent to another facility for a certified radioassay to support disposal of the waste at the WIPP in New Mexico.

The final project step in the process is the flowdown of technical and regulatory requirements. Technical requirements are contained in "*Nondestructive Assay Management Program,"* [2] and "*Specification for Mobile Assay for Suspect TRU Waste in the Low-Level Burial Grounds,"* [3]. Regulatory requirements specific to classification of waste for disposal have been developed through a Sample Analysis Plan [4] and DQO process [5]. The DQO process defines the required decisions and data needed to disposition the waste. In addition, various daily quality controls checks are routinely performed to ensure the radioassay system is operating properly.

Identification of Unknown Waste Containers

Unknown wastes are defined as waste containers that are removed from the burial grounds without legible labels or markings such that the contents of the container cannot be identified or associated with its corresponding records or acceptable knowledge data package. Identification and efficient resolution of unknown waste containers is important to minimize project costs for handling unknown conditions. Research and identification of an unknown waste is analogous to an archeological excavation using carbon-14 dating to determine information about the site. In most cases, a simple records search and comparison can be used to identify or resolve an unknown waste. In more complex situations radioassay data can be used to resolve unknown waste containers.

A module usually contains 12 drums across the front, 12 drums wide and 4 drums high (see Fig. 1) or a total of 576 drums. In the simple case, one drum might be an unknown waste container. This scenario is easily resolved by matching the burial records against the unknown container in the module. When a module is found with multiple unknown containers, in some cases a process of elimination is used to match the container coordinates, location, or burial date against the records. Knowing that a group of drums was received and buried on the same date is often enough to identify the unknown container. These simple techniques have resolved many unknown containers.

The more complex case exists when multiple drums have illegible or no markings and the process of elimination does not resolve the unknown waste. In this case, it is possible to use radioassay results to associate the unknown drum with the proper burial records. For example,

one module has three unknown containers and there are three burial records left after all other containers have been removed. The burial records and associated accountability records indicate the drums contain less than 1 gram, 10 grams, and 40 grams plutonium, respectively. Radioassay results confirm the three unknown drums contain 1E-03 grams, 15 grams and 35 grams plutonium, respectively. It is then reasonable to match the radioassay results with corresponding burial records based on gram quantity and resolve the unknown waste containers.

Finally, the situation may exist where the total plutonium activity in a waste as determined by radioassay and the process of elimination does not resolve the identity of an unknown waste container. In this case, further review of the radioassay results may be used to identify the waste generator. As noted and described in the next section, numerous on-site and off-site waste generators have used the Hanford Site for the disposition of suspect TRU waste. With a few exceptions, most of these waste generators processed radioactive material with a standard isotopic mixture. For example, the Hanford Plutonium Finishing Plant (PFP) typically processed plutonium at 6 and 12 % plutonium-240. In cases where the isotopic mixture can be identified it may be used to identify an unknown waste container.

In some cases, due to conflicting radioactivity and high activity levels in the waste, assay results are indeterminant for the purpose of segregating TRU from LLW. Fig. 2 depicts a gamma spectrum where it is not possible to discern the TRU radionuclide concentration below 100 nCi/g. In this specific case, this drum contained excessive cesium-137 and some cobalt-60 which masked the plutonium signal. The blue vertical line in Fig. 2 is a marker corresponding to the 413.71 keV plutonium-239 peak. The red curve in Fig. 2 depicts the gamma spectrum for this waste drum. The point where the blue and red lines intersect corresponds to a minimum detectable activity for Pu-239 at a TRU concentration greater than 100 nCi/g. The drum may be LLW but it is not possible to see the actual TRU concentration or plutonium activity at a level below 100 nCi/g to verify a LLW classification. Indeterminant assays are handled as TRU waste and sent to another facility for further nondestructive analysis and processing.

During retrieval of suspect TRU waste from the WRP, over 12,000 drums have been removed from the burial trenches. Forty-nine of these drums identified as unknown drums have been resolved by the techniques described above. This avoids the cost of characterizing the unknown waste, and these containers are now processed as part of their respective waste streams.

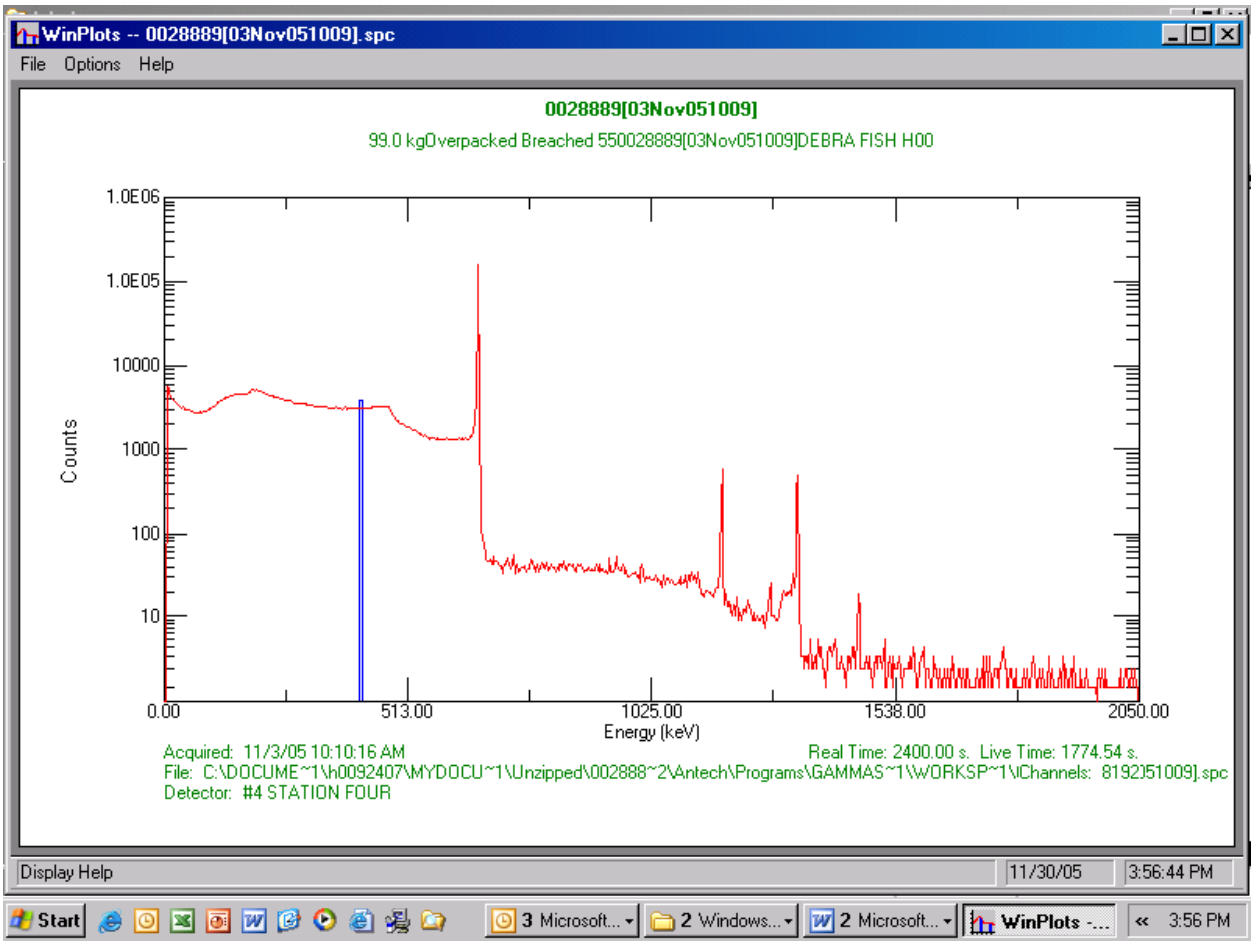

Fig. 2. Radioassay gamma spectrum showing indeterminant results

On-site and Off-site Waste Generators

Suspect TRU waste was received from many different on-site and off-site waste generators. Documentation on each waste generator is compiled into an Acceptable Knowledge (AK) data package. Each waste generator has certain of its history that can be traced or identified through radioassay. The next section provides greater detail and describes historical perspective on five of the waste generators, based on the gamma signature of the suspect TRU waste. Table I provides a list of suspect TRU waste generators that are associated with the WRP.

Table I. Hanford Suspect TRU Waste Generators

Facility Code	Waste Generating Source
308 Facility	Plutonium Fabrication Pilot Plant (Plutonium Laboratory and Fuels Development Laboratory)
318 Facility	High Temperature Lattice Reactor
324	Chemical Engineering Laboratory
325	Radiochemistry Building
340	Retention and Neutralization Complex
105KE	Battelle Northwest Laboratory
105N	Plutonium production reactor, reactor operation experimentation and domestic power production
1706K	Plutonium production reactor, reactor operation experimentation
200W	Unspecified locations in 200 West Area
202A / 202AL	Plutonium Uranium Extraction Plant (PUREX)
209E	Critical Mass Laboratory
216Z9	PFP Complex Building and Crib
222S	Reduction Oxidation (REDOX) Control Laboratory
231Z	Isolation Building (Concentration Building) Plutonium metallurgical laboratory
233S	Plutonium Concentration Facility
2345Z	Plutonium Finishing Plant (PFP)
2WTF	West Tank Farms
327 / 327C	Radiometallurgical Building
BABCX	Babcox and Wilcox
BATCO	Battelle Columbus laboratory
BETTS	Bettis Atomic Power Laboratory
CUPRC	Center for Energy and Environmental research
ESG	Rockedyne Energy Systems Group
EXXON	Exxon Nuclear Systems
LBLAB	Lawrence Berkeley laboratory
MCGEE	Kerr-McGee, Cimarron Plutonium Fuel Fabrication Facility
VAL	General Electric Vallecitos Nuclear Center
WARD	Westinghouse Advanced Reactor Division

Radionuclide Signatures

The gamma spectrum for a subset of these waste generators from Table I is presented as a signature or method for relating the waste to the waste generator. This section provides greater detail and describes historical perspective on five of the waste generators based on the gamma signature of the suspect TRU waste. The gamma spectrum is presented by plotting counts versus energy.

Plutonium Isotopic Signature

Plutonium isotopic mixtures can range from approximately 1 % Pu-240 through 6, 12, and 23 % Pu-240. Initially, uranium was irradiated for very short periods to produce super grade weapons plutonium containing 0.9 % Pu-240. Later, weapons programs were optimized to produce weapons grade plutonium at 6 % Pu-240. Fuels grade plutonium at 12 % Pu-240 was produced for breeder and other reactor programs. Research grade plutonium contains greater than 12 %

Pu-240 and is frequently found at 18 to 23 % Pu-240. Table II provides the standard 20-year decayed plutonium mass distributions for 6, 12, and 23 % Pu-240 [6]. These mixtures can be used to identify the origin of the waste.

Table II. Standard 20-Year Decayed Plutonium Mass Distributions (weight %)

Isotope	Nominal 6% Pu-240	Nominal 12% Pu-240	Nominal 23% Pu-240
Pu-238	0.03%	0.08%	0.09%
Pu-239	93.21%	83.95%	70.60%
Pu-240	6.02%	12.97%	23.86%
Pu-241	0.22%	1.10%	1.60%
Pu-242	0.02%	0.03%	1.30%
Am-241	0.50%	1.75%	2.55%

Generally, the isotopic ratio used during radioassay is based on the AK data package from the nuclear material accountability records. Accountability records include the nuclear material item transfer or nuclear material transaction report (DOE/NRC Form 741). When enough plutonium is present in the waste container, radioassay is used to determine or confirm the isotopic ratios. However, for the WRP, the drums planned for assay (those with records showing 1 gram plutonium or less) seldom have sufficient plutonium and the isotopic ratios are based on the records. In the case where the ratio is not known the next higher ratio is used. For example, waste containers with records indicating 6.21 % to 13 % Pu-240 are assigned the isotopics for 12 % Pu-240. For a facility like the Plutonium Finishing Plant which is known to process 6 % and 12 % material, an unknown drum would be assigned the 12 % Pu-240 isotopic composition. This effectively over estimates the TRU radionuclide concentration in the waste container and helps to ensure a TRU waste is never mis-classified as LLW.

Thorium-232 and Uranium-233

Thorium-232 is a naturally occurring radioactive isotope. There are many uses for thorium including the development of a thorium-232 uranium-233 fuel cycle. During 1965-1966, an experimental processing of commercial thorium nitrate into thorium oxide powder was carried out in the Hanford Uranium Oxide (UO3) Plant, using the old electric pot calciners. The goal of this work was to produce thorium oxide powder suitable for fabrication into reactor target elements for U-233 production [7,9]. However, for reasons unrelated to the UO3 Plant, the use of thorium oxide powder was abandoned at the Hanford Site, in favor of experiments with thorium wafer targets [9,10]. Th-232 is identified through radioassay by ANTECH[1] by the 238.6, 583.3, and 911.3 keV peaks. Thorium materials have been handled through many nuclear processes including experimental reactors. The most likely waste generator of this type of waste is Battelle Northwest Laboratory.

Argonne National Laboratory sent suspect TRU waste to Hanford from 1985 thru 1987. This particular waste stream contained significant quantities of thorium-232 and uranium-233 from the development of the Light Water Breeder Reactor – Proof of Breeding Program. The waste is from research and development on the breeder reactor program and is remote handled with about 300 curies of mixed fission products per container. This waste stream does not contain TRU isotopes above the 100 nCi/g definition for TRU waste.

[1] ANTECH Corporation is the radioassay contractor.

Neptunium-237

Fig. 3 depicts a gamma spectrum from a drum showing a significant quantity of neptunium-237. Np-237 is identified by the 300.14 and 312.17 keV peaks. Original records for this particular drum show 14 grams Np-237 and enriched uranium at 99.8 % U-235. Radioassay identified 3 grams Np-237 and 9 grams of U-235. These isotopes are readily discernable in the gamma spectrum. Methods for producing and separating Np-237 are described in *Hanford Reactor and Separations Facility Advantages* [9]. Np-237 was separated at Plutonium Uranium Extraction (PUREX) Facility and processed through the glovebox in Q cell. The waste depicted in Fig. 3, generated from the High Temperature Lattice Test Reactor, 318 Facility, by Battelle Northwest Laboratory, is described as plastic bags containing miscellaneous slip-lid metal cans and plastic bags with source material randomly distributed. This drum is identified as TRU waste and will be certified for disposal at the WIPP.

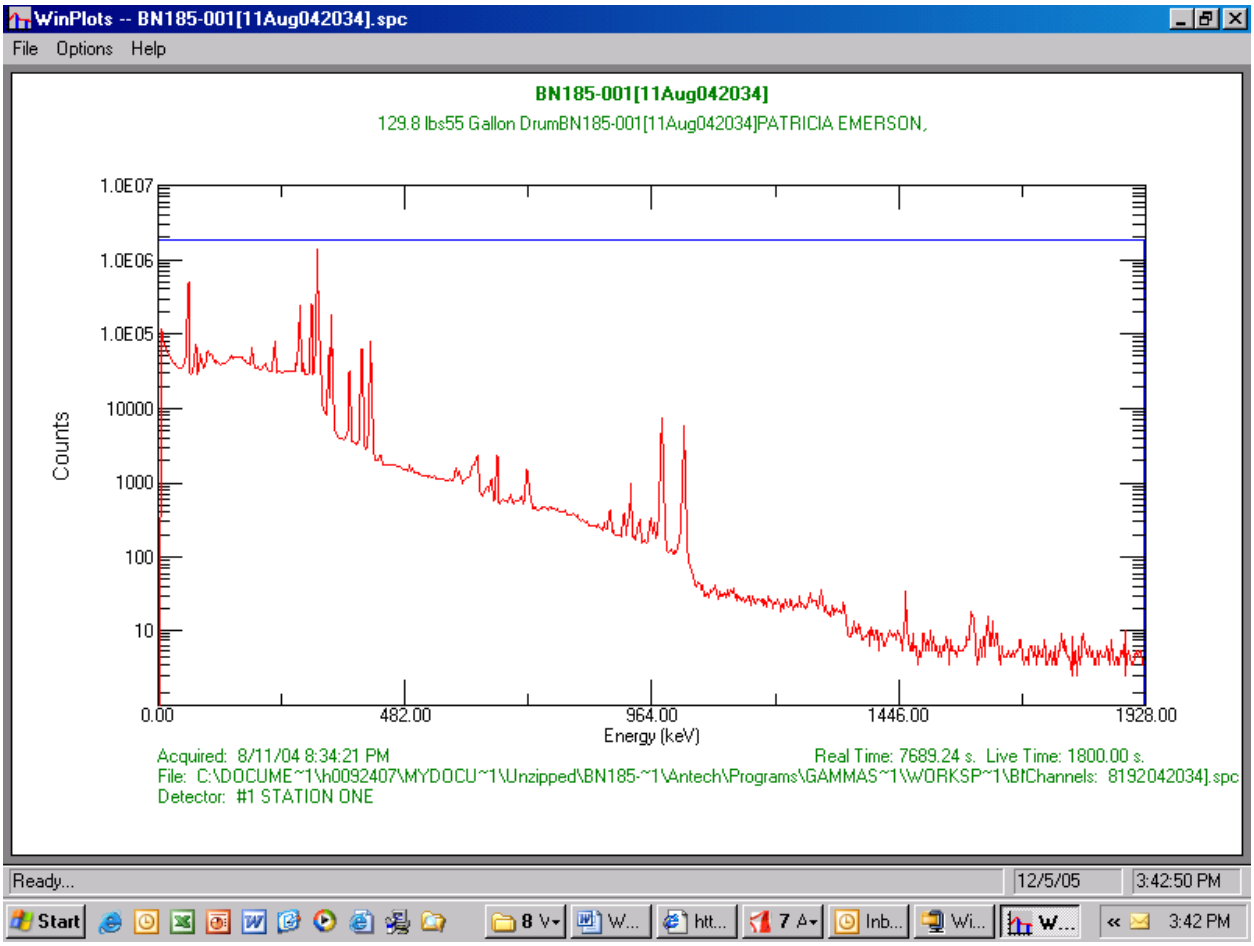

Fig. 3. Radioassay gamma spectrum showing Np-237, U-235 and Cm-243 gamma peaks

Promethium-146

Promethium-146 is an unusual isotope to see in the suspect TRU wastes and clearly marks this waste as originating from the research facilities at Battelle Northwest Laboratories. Fig. 4 shows a gamma spectrum for a TRU drum from the 325 Radiochemistry Building. The blue vertical line in Fig. 4 is a marker corresponding to the 413.71 keV plutonium-239 peak. This drum is

classified as TRU waste at about 1600 nCi/g based on the plutonium-239 peak. Promethium-146 is identified in the waste drum by the 747.24, 735.93, 453.88, and 633.25 keV peaks and two low abundance peaks (589.8 & 146.2 keV) in the gamma spectrum. This drum also contains curium-244. Promethium is a rare-earth element produced by fission of uranium or by neutron bombardment of neodymium. It is not naturally occurring and was first separated from man-made products in 1963. Promethium was also recovered from Shippingport Reactor Fuel reprocessing wastes at the Hanford Site [8,9].

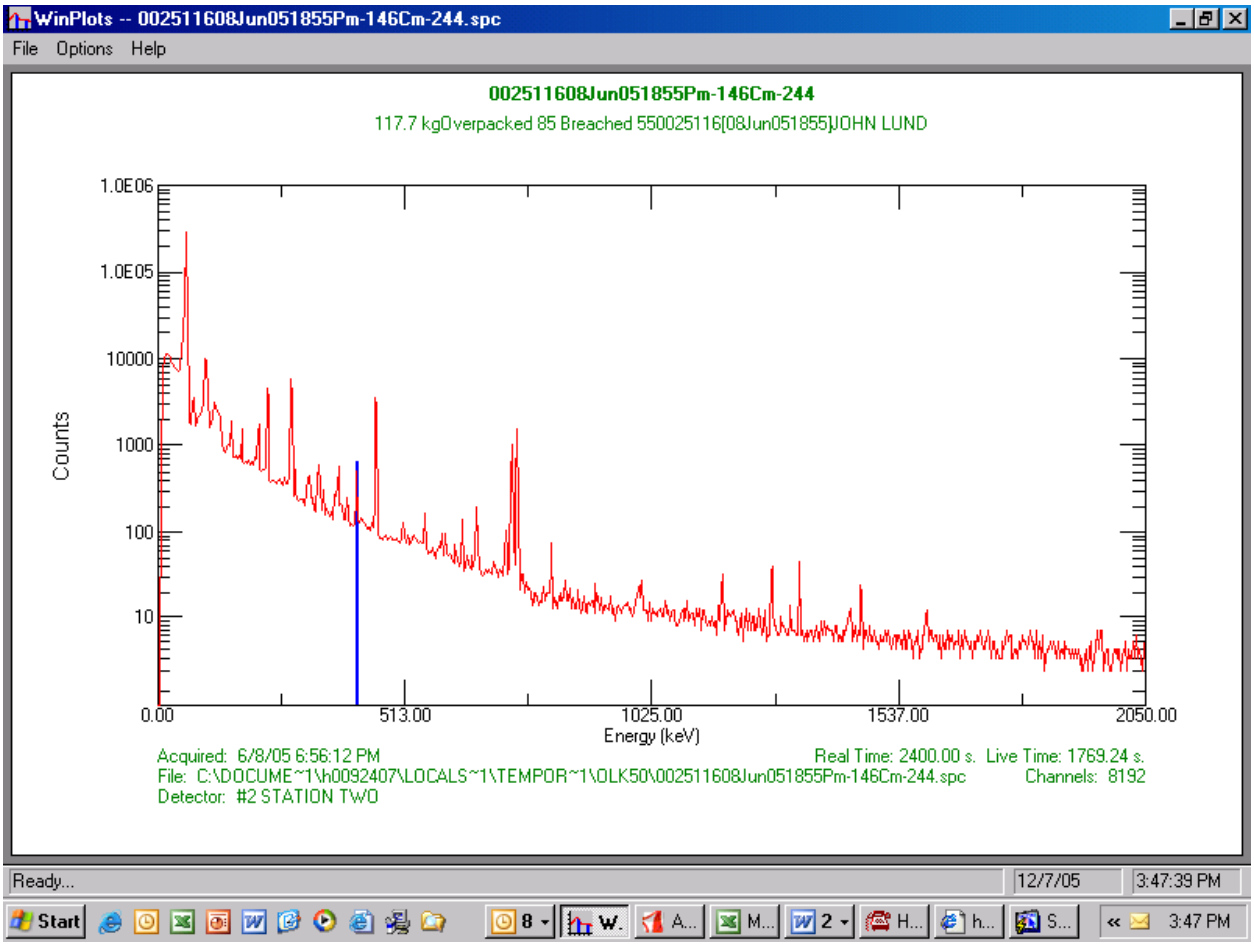

Fig. 4. Radioassay gamma spectrum showing Pm-146 and Cm-244 gamma peaks

Curium and Plutonium-238

Curium-242, curium-244, and plutonium-238 are heat source materials used in the space programs. These isotopes were produced and separated in research facilities and subsequently used in compact thermionic and thermoelectric power generators. Curium isotopes and elevated levels of plutonium-238 are readily identified in waste containers by radioassay. The 99.6, 152.2, and 766.4 keV energy peaks are used to identify and quantify plutonium-238. Methods for producing and separating curium and Pu-238 are described in *Hanford Reactor and Separations Facility Advantages* [9]. Other curium isotopes exist, and Fig. 5 depicts the gamma spectrum for a drum containing americium-241, plutonium-238 and curium-243. Energy peaks at 228.19 and 277.6 keV are used to identify the curium-243. The blue vertical line in Fig. 5 is a

marker corresponding to the 413.71 keV plutonium-239 peak. This drum contains glovebox wastes and originated from the 340 facility managed by the Battelle Northwest Laboratories.

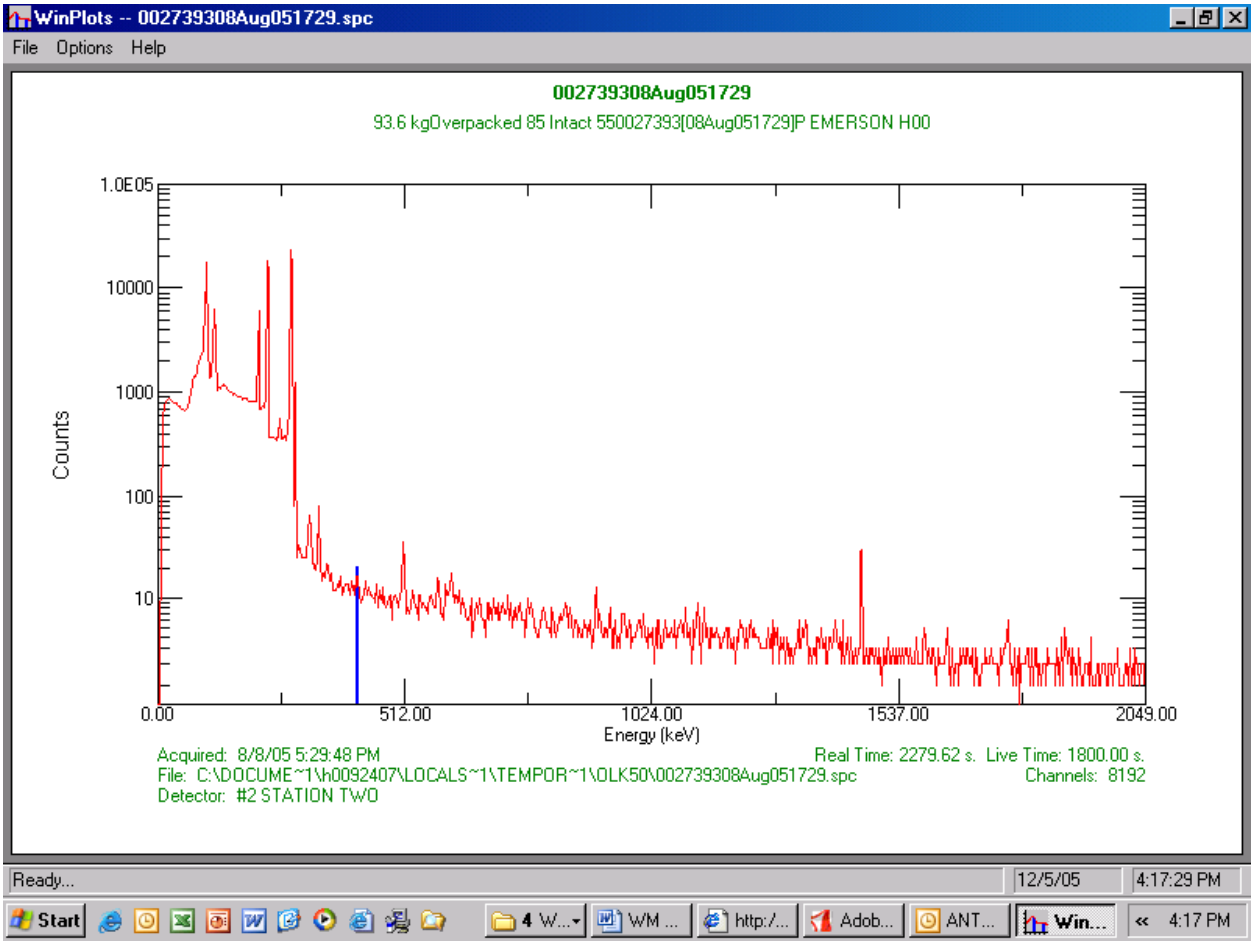

Fig 5. Radioassay gamma spectrum showing Am-241, Pu-238 and Cm-243 gamma peaks

Cobalt-60 and Cesium-137 Sources

Fig. 6 depicts the gamma spectrum for a suspect TRU drum from the Westinghouse Hanford Company, 340 Facility. Normally, these drums will exhibit the characteristic gamma spectrum for plutonium at 6 % or 12 % Pu-240. The blue vertical line in Fig. 4 is a marker corresponding to the 413.71 keV plutonium-239 peak. In this case there are no plutonium peaks identified above the minimum detectable activity, so this drum is classified as LLW at 74 nCi/g based on the plutonium MDA values. However, the Cs-137 and Co-60 peaks are readily identified as the only significant activity in this drum, and it appears that small radioactive sources were disposed in the waste. The non-TRU waste stream is treated and disposed of on-site, according to regulatory requirements. As an example of treatment, one mixed LLW stream is super-compacted and macroencapsulated (grouted) in a waste box for disposal on-site.

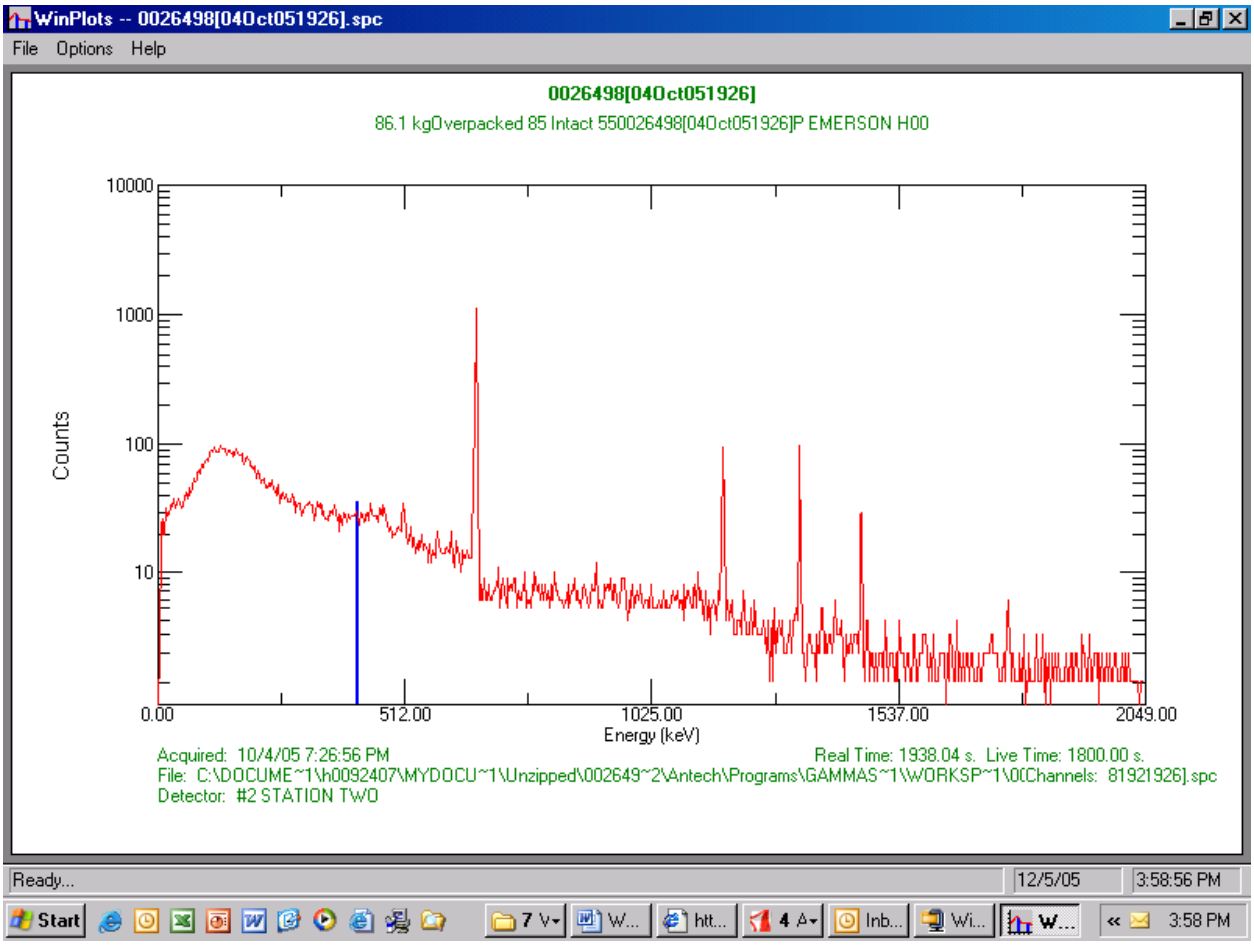

Fig 6. Radioassay gamma spectrum showing Cs-137 and Co-60 gamma peaks

FUTURE PROJECT CHALLENGES

Current waste retrieval activities are centered in the 218-W-4C burial ground. At the start of the WRP the 218-W-4C burial ground had approximately 3,732 m^3 of suspect TRU waste in drums and 3,359 m^3 suspect TRU in other containers such as metal and fiber reinforced plastic boxes. Three other burial grounds are planned for retrieval and removal of the suspect TRU waste. The 218-E-12B burial ground contains approximately 638 m3 of suspect TRU from the Hanford's PUREX Facility waste generator buried between 1970 and 1972. Due to fission products in the PUREX waste stream, this burial ground will require the use of a combination of gamma and neutron radioassay detector systems in order to adequately characterize the waste. The 218-W-3A burial ground contains approximately 4,104 m^3 suspect TRU waste buried between 1970 and 1984. This burial ground presents challenges for retrieval due to the age and degradation of the waste containers and waste form (lab packed liquids etc.). The final burial ground 218-W-4B for retrieval contains approximately 3,219 m^3 suspect TRU waste. This burial ground presents challenges due to the unique manner in which drums were stacked in a "V" arrangement. Some of the older waste is identified in the burial records as remote handled waste and is not suitable for current radioassay techniques. Overall, the major challenge confronting radioassay is throughput capability to assay up to 5,000 containers per year.

CONCLUSION

A project management approach is applied to radioassay activities in the WRP to ensure effective and efficient management of scarce resources to classify suspect TRU waste. Radioassay provides a snapshot of the various waste generators and varied history of the Hanford Site. In addition to meeting the primary DQO to classify suspect TRU waste as TRU or LLW, radioassay can be used to help identify unknown wastes. Anytime an unknown can be resolved it eliminates or avoids the very costly expense to reprocess the waste to characterize the waste contents. Radioassay may be used to identify unknowns by identifying the quantity of plutonium in a waste container. Other methods used to identify an unknown waste include matching the gamma signature to the documentation from AK. Unique or unusual gamma signatures are tools in a project management arsenal used to identify the origin of a particular waste to aid in the efficient cleanup of the Hanford Site. This process is analogous to an archeological site using Carbon-14 dating to identify information about the contents of the site.

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