RADIOASSAY AT THE INEEL DURING THE 3100 M^3 PROJECT

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ABSTRACT

The Radioactive Waste Management Complex (RWMC) at the Idaho National Engineering and Environmental Laboratory (INEEL) is being used as a temporary storage facility for transuranic (TRU) waste generated by the United States nuclear weapons program. There has been, and continues to be, a very large effort to characterize, certify, and ship this legacy waste, in compliance with all applicable regulations, to the Waste Isolation Pilot Plant (WIPP) in Carlsbad, New Mexico for permanent disposal. Nondestructive assay (NDA) was used to characterize TRU waste for shipment/disposal to WIPP during the 3100 m^3 Project. This paper describes the NDA systems, summarizes NDA history at the RWMC, and summarizes the lessons learned during the 3100 m^3 Project.

In 1995 the State of Idaho, the Department of Energy, and the United States Navy signed a multifaceted legal document requiring Department of Energy to remove all forms of radioactive waste from the INEEL. One aspect of the agreement is to remove 3100 m^3 of TRU waste prior to January 1, 2003 from RWMC. In October of 1996 congress allocated funding for the 3100 m^3 Project to fulfill the agreement.

Prior to 3100 m^3 Project TRU waste radioassay research and development was performed at the INEEL (1980s - 1990s). The preliminary assay operation years covered in this paper are 1993 – 1997. During the preliminary assay operation years the NDA efforts relied on a Passive/Active Neutron assay system coupled with a gamma spectrometry system. Initial radioassay efforts saw the:

- Determination of the relative isotopic composition of each container.
- Development of a total uncertainty measurement method.
- Estimation of the total uncertainty of each measurement.
- Development and integration of a gamma-ray based isotopic mass ratio method.

The WIPP certified production radioassay commenced in late September of 1997. Production assay operation continued through the completion of the 3100 m^3 Project in late 2002. The state of the NDA systems and methodologies at the close of the 3100 m^3 Project reflects a long list of applied knowledge and experiences:

- No one assay technique that can be applied to all waste forms.
- The waste assay technique must be appropriate to the waste form and waste matrix.
- Knowledge of the waste form and packaging is absolutely necessary and should be as extensive as possible.
- Limitations of each assay technique must be well known.
- Calibration requirements need to be flexible enough to take advantage of advanced and different NDA methods.
- Calibration efforts must be rigorous and well documented.
- Throughput requirements should be well defined.
- Accuracy and precision goals should be well defined.
- Sensitivity goals should be well defined.
- Quality assurance methods should be well defined.
- Software and Program Documentation should be considered "Living Documents".
- Automation of functions was the most desirable solution to "Special Cases".
- Advantageous to have well trained and experienced personnel.

INTRODUCTION

The Radioactive Waste Management Complex (RWMC) at the Idaho National Engineering and Environmental Laboratory (INEEL) is being used as a temporary storage facility for transuranic (TRU) waste generated by the United States nuclear weapons program. There has been, and continues to be, a very large effort to characterize, certify, and ship this legacy waste, in compliance with all applicable regulations, to the Waste Isolation Pilot Plant (WIPP) in Carlsbad, New Mexico for permanent disposal. Nondestructive assay (NDA) was used to characterize TRU waste for shipment/disposal to WIPP during 3100 m^3 Project. The NDA equipment that performed radioassay for characterization was housed at the Stored Waste Examination Pilot Plant (SWEPP) at the RWMC. Three distinctly different radioassay measurement systems were in use over the life of the 3100 m^3 Project. This paper describes the three NDA systems, summarizes NDA history at the RWMC, and summarizes the lessons learned during the 3100 m^3 Project.

The effort to ship TRU waste to WIPP is being driven by the Idaho Settlement Agreement [1] which is a multifaceted legal document requiring the Department of Energy (DOE) to remove all forms of radioactive waste from the INEEL. One aspect of the agreement is to remove 3100 m^3 of TRU waste prior to January 1, 2003 from the RWMC. This agreement between the State of Idaho, the Department of Energy, and the United States Navy was signed in October of 1995. Funding for this project was allocated in October of 1996 by congress. Prior to this time, work to characterize TRU waste was performed under the WIPP Experimental Test Program (WETP). The WETP project started in the early 1980's and continued until the creation of the 3100 m^3 Project. The court-approved milestone was achieved on September 30, 2002.

RADIOASSAY SYSTEMS

RWMC used 3 different measurement systems over the life of the 3100 m^3 Project to perform NDA. A Passive/Active Neutron (PAN) assay system coupled with a high-resolution gamma spectrometry system was used initially. The high-resolution gamma systems were the SWEPP Gamma-ray Spectrometer (SGRS) and Waste Assay Gamma Spectrometer (WAGS). In their support roles to the PAN assay system, the gamma systems determined the relative isotopic material composition data, which were combined with the PAN response data to determine absolute estimates of the radionuclide inventory and compute the associated derived quantities; total alpha activity, TRU alpha activity, fissile gram equivalent. During the last year of the 3100 m^3 Project, SGRS and WAGS were also used, independent of the PAN assay system, as (standalone) absolute assay systems.

As a whole, the three NDA systems assayed 19,669 distinct containers during the production period of the 3100 m^3 Project. Based on radioassay requirements 83% of the drums assayed by an NDA system were acceptable for shipment to WIPP.

PAN/Gamma

In 1983 Los Alamos National Laboratory (LANL) delivered a PAN assay system to the INEEL, the system was classified as a second-generation PAN assay system. It was a successor to the 1st generation PAN assay system, which was developed for safeguards measurements. Whereas, the 2nd generation PAN assay system was adapted to measure TRU waste. There were a number of 2nd generation PAN assay systems produced by LANL and delivered to different DOE facilities. Each PAN assay system was slightly different in design and construction, and therefore had to be treated as a one of a kind system. The closest systems in design to the INEEL PAN assay system were delivered to the Savannah River Site and the Rocky Flats Environmental Technology Site (RFETS). The PAN assay system gave RWMC the capability to assay 55-gal TRU waste drums. The TRU waste sent to WIPP during the 3100 m^3 Project from INEEL was received from RFETS. Prior to receipt of the PAN assay system, INEEL relied solely on the transportation records and special nuclear material records that were received by INEEL from RFETS to determine the radionuclide content of a container. The PAN assay system was used from the start of the radioassay program at the RWMC through the life of the 3100 m^3 Project.

A complete description of the PAN assay system is given in the engineering design file (EDF)-606 [2]. The major structure is an enclosure that surrounds the drum on all four sides, top, and bottom and can be classified as a 4π detector. The outer skin of the enclosure is constructed of aluminum sheets, inside is housed a layered arrangement

of graphite, non-borated and borated polyethylene, and cadmium wrapped and bare helium-3 neutron detectors. In the PAN assay system design, the shielding around the different detector banks was arranged such that one set of detector banks were surrounded by cadmium, and borated rubber, and moderator and were sensitive to fast neutrons only. The second set of detector banks were surrounded by moderator only and were sensitive to neutrons of all energies. The first set of detector banks was called the "shielded detector banks" and the second set was called the "bare detector banks".

The PAN assay system operated in both passive and active modes, both of which were needed to quantify the final assay values. The passive mode was sensitive to neutrons produced by spontaneous fission, from Pu-240 primarily, and (α,n) interactions in the waste matrix. The active mode was sensitive to neutrons produced by thermal neutron induced fission.

In the passive mode, coincidence counting techniques were used to differentiate neutrons produced by spontaneous fission from uncorrelated neutrons produced by (α,n) interactions and cosmic background. Initially, the PAN passive coincidence mode operated using "one shot" coincidence gates [3]. Two coincidence gates were used; the long-gate mode and the short-gate mode. The long-gate mode operated with a time gate window of 250 μs and detected coincidence events originating from all the detectors in the enclosure, both bare and shielded. The short-gate mode operated with a time gate window of 45 μs and detected coincidence events originating from only the shielded detectors in the enclosure. The use of two coincidence modes was developed to extend the dynamic range of the PAN passive mode from a few grams of plutonium to at least 200 g of plutonium. Originally, the PAN assay system's analysis software selected the short-gate coincidence mode results if that mode's coincidence count rate determined a measured plutonium mass of greater than 20 g. The long-gate coincidence mode results were used if that mode's coincidence rate produced a measured plutonium mass that was between 10 g and 20 g. The choice of passive mode results was changed to whichever passive mode results had the smallest relative uncertainty on the plutonium mass and the algorithm change was made in the early 1990's. Practical experience also showed that the one-shot coincidence methods would underestimate the mass of plutonium due to detector pulse pileup and dead time losses. As a result, in 1996 a shift register coincidence (SRC) system, which virtually eliminates dead time losses, was added to the PAN passive mode acquisition hardware.

The active mode of the PAN assay system is based on the differential dieaway technique [4]. This mode used a deuterium on tritium (D-T) neutron generator to produce a series of bursts of 14 MeV neutrons which moderated to thermal energies and interacted with Pu-239, U-233 or U-235 to produce fission that in turn produced 2 or more fast fission neutrons. As a result, the neutron levels (and consequently the neutron detector count rates) inside the PAN assay system fall off (or die away) after each D-T generator pulse with an exponential dieaway. Two time-gate windows were set following each neutron generator burst. The first time-gate window was set to accumulate counts when the interrogation neutrons were thermalized and most likely able to produce fission is referred to as the active mode early gate. The second time gate window, referred to as the active mode late gate, was set when all the interrogation neutrons have died away and only background and (α,n) neutrons would be detected. The early gate time window was set to be from 700 μs to 2,700 μs and the late gate time window was set to be from 5,700 μs to 15,700 μs. The late gate count rate was used to correct the early gate count rate for background and (α,n) neutrons. The net count rate was then used to derive the active mode plutonium mass. By this approach the active mode automatically corrected for background.

Because the active mode used an interrogating neutron flux, it was thought to be inherently more sensitive than the passive mode. Consequently, the active mode assay results were selected as the reported results when the plutonium mass was small. Originally the decision point between active and passive results was based on the active plutonium mass being 10 g. That is, when the active plutonium mass was above 10 g the passive assay results were reported and when the active plutonium mass was below 10 g the active assay results were reported. In 1996, the decision criterion for reporting active mode results versus passive mode results was changed such that the active mode results were reported if the passive mode plutonium mass plus 2σ (1.95 times its counting error) were less than 5 g.

The INEEL PAN assay system, operated in conjunction with a gamma system (operating in an isotopic-mass-ratio mode), that was used to quantify the mass values for the following nuclides: Am-241, Pu-238, Pu-239, Pu-240, Pu-241, U-233, U-235, and U-238. The PAN assay system directly measured a fissile signal due to Pu-239 or U-235 fission (active mode) or Pu-240 spontaneous fission (passive mode). A gamma system, either the SGRS or the

WAGS was used to supplement the PAN measurements by providing the relative mass ratios of Pu-238/Pu-239, Pu-240/Pu-239, Pu-241/Pu-239, Am-241/Pu-239, Am-241/U-235, U-235/Pu-239, U-233/Pu-239 and U-235/U-238. The gamma system analysis code for mass ratios analysis was called the SWEPP Gamma Analysis Package (SGAP) [5]. The mass ratio data and the PAN data were combined using the SWEPP Assay System (SAS) analysis code [6] to produce isotopic mass values for the nuclides listed above. This combined assay method is referred to in this document as PAN/Gamma assay system.

The PAN/Gamma assay system was in operation at the start of the project (September of 1997) and continued to assay waste containers until it was placed in standby mode (June of 2002). From the start of the project and until December of 2000, the PAN assay system operated in conjunction with SGRS. In mid-December of 2000, the WAGS system was placed into operation. The PAN instrument could then be provided mass ratio data from either the SGRS or the WAGS to support the PAN measurements. The PAN/Gamma assay system was certified to ship both debris and sludge waste to the WIPP [7,8,9,10].

The PAN/Gamma assay system operated prior to implementation of the Contact-Handled Waste Acceptance Criteria (CH-WAC) [11] under the WIPP Waste Acceptance Criteria (WAC). The WIPP WAC required daily quality control checks (PAN), daily/weekly calibrations on the Gamma system, replicate/batch, annual calibration conformation, and Performance Demonstration Program (PDP). The quality control events will not be addressed in this report. The technical procedure (TPR) used for the daily operation of the PAN assay system was TPR-1573 [12] while the gamma system was operated under TPR-1588 [13].

SGRS Absolute and SGRS Mass Ratio

The SGRS system was the first permanent gamma system placed in service. Prior to the SGRS system, temporary gamma systems were used to perfect the isotopic measurement method. The SGRS system had four high-resolution high-purity germanium (HPGe) gamma-ray spectrometers and a shielding enclosure with 15.24 cm thick pre-World War II steel walls. The SGRS system's enclosure had 4 penetrations in the shield to position 4 HPGe detectors. A waste drum was placed on a drum rotator inside the enclosure by the forklift; the drum rotator was used to rotate the drum while gamma spectra were accumulated [14].

An Ethernet communication link was used to communicate between the detector acquisition hardware and gamma acquisition computer. Initially, the gamma acquisition computer was a VAX station 4000 Model 60 computer. The VAX based system was later replaced by a Windows NT based personal computer (PC). Each HPGe gamma spectrometer was connected to a high voltage power supply, a low-noise preamplifier with dual-energy pulser interface, a linear amplifier, and an analog-to-digital converter, which was modified to process dual-energy pulser signals to and from the preamplifier and an acquisition interface module.

The initial software package used to acquire and analyze SGRS gamma spectrum data consisted of three modules [15]:

- Gamma spectrum acquisition and control program (GCS).
- VAX Gamma-ray analysis program (VAXGAP).
- Actinide mass ratio calculation program (BGAV).

The main core of this software package was VAXGAP [16,17]. The program's long pedigree resulted from extensive testing, use, and national validation and verification in the Radiation Measurements Laboratory at the INEEL, Test Reactor Area.

The SGRS software package went through three major changes over the life of the NDA project.

1. A method was developed to successfully sum gamma spectra from different HPGe detectors with different energy calibrations without a loss in energy resolution. The isotopic mass ratio calculations in the SGRS software were changed to take advantage of the new spectrum summing method. This change was implemented in BGAV3 and it greatly improved the accuracy and consistency of the SGRS mass ratio results [15].

2. Due to the change of the SGRS computer from a VAX to a PC in 2001, a completely new software package was developed and implemented. The new package was called the SWEPP Gamma Analysis Package (SGAP) [18]. Like its predecessor this software package consists of three major modules:

    - The acquisition and control module (GCPC).
    - Gamma spectrum analysis module (PCGAP).
    - Isotopic mass ratio calculation module (SAAC).

3. In May of 2002, an absolute assay analysis module developed at INEEL was added to the SGRS software package. The absolute assay package was called SWEPP Absolute Analysis Package (SAP) [19] and it allowed the SGRS to operate as a standalone absolute assay unit, separate from the PAN/Gamma operation. As with SGAP, SAP had three modules, the first two modules were the same modules that were used in SGAP:

    - Acquisition and control module (GCPC).
    - Gamma spectrum analysis module (PCGAP).
    - Isotopic mass and derived quantities calculation module (SRAC).

The absolute assay operation of the SGRS system did not require any modifications to the hardware or the acquisition/gamma-spectrum-analysis software. The only difference between mass ratio mode and absolute assay mode was the analysis performed at the tail end of the process. To the SGRS system operator the only difference was the selection of a different icon on the computer monitor. This meant that the SGRS system could be switched from mass ratio mode to the absolute assay mode (and vice versa) with no time delays to reconfigure the hardware or to recalibrate. SGRS system was totally compatible for either mass ratio operation to support PAN assay or for standalone absolute assay operation. However in order to distinguish between the two modes, the SGRS system when operated as an absolute assay system is referred to as SGRS Absolute in this paper.

SGRS Absolute detected and directly measured the mass concentrations of Am-241, Pu-238, Pu-239, Pu-240, Pu-241, U-233, U-235, and U-238. Certification included both debris and sludge waste types [10]. This instrument was placed into operation under the requirements of the CH-WAC. Therefore, the system was required to have a daily quality control event, background event, a weekly interfering matrix test, and participate in an annual PDP test. The quality control events will not be addressed in this report. The daily operating procedure, TPR-1588 [13], was modified to include operating the SGRS in either the isotopic mass ratio mode or the absolute assay mode.

WAGS Absolute and WAGS Mass Ratio

The WAGS system was originally purchased to perform the same functions as the SGRS system; provide isotopic mass ratio data (in parallel with the SGRS) to support the PAN analysis. The WAGS system operated as a second isotopic gamma system in support of the PAN assays to reduce the backlog produced by the gamma spectrum acquisition. It was placed into operation at SWEPP in December of 2000.

The WAGS system was designed to analyze the radionuclide content of a 55-gal drum using a combination of six HPGe gamma-ray detectors. The housing for the WAGS system was a Canberra Q-2 rectangular enclosure with 10.16 cm thick low-background steel shielding. Penetration holes in the shield provided for six HPGe detectors and three transmission sources. A drum transport system moved drums in and out of the system and a rotator/lift assembly lifted and rotated the drum in the enclosure during data acquisition. This system was fully automated in that a maximum of six drums were loaded onto the conveyor and sequentially moved through the assay system. Operator interface other than data entry was not required [20].

When operating as an isotopic gamma system in support of the PAN assay, the WAGS data acquisition hardware and software were duplicates of those used on the SGRS system. The major exception was that only three detectors were used in the WAGS system and four detectors were used in the SGRS system. This meant that the WAGS system did not use all six detectors when it operated in the isotopic mass ratio mode.

In 2002, the WAGS system was converted to operate as a standalone absolute assay system, independent of the PAN assay system. To distinguish absolute assay operation from its isotopic mass ratio operation the term WAGS Absolute is used when WAGS operated as an absolute assay system. The reconfiguration required both changes in hardware and software. Under the absolute assay operation, the WAGS transmission source housings contained three 5 mCi Ba-133 radioactive sources and the shield doors would open to allow diametric attenuation measurements of the drum contents. As a standalone absolute assay system, a PC with an OS/2 operating system controlled the WAGS system. Canberra Industries provided the acquisition and analysis software; i.e., Genie PC and Gamma Waste Assay System. During an assay, energy dependent attenuation coefficients were calculated based on the analysis. These attenuation coefficients and/or weight based density coefficients were used in conjunction with the gamma-ray spectral analysis to determine the radionuclide content of a drum [20].

WAGS Absolute was placed into operation in mid-May of 2002 and operated through the end of the 3100 m^3 Project. During this time 3000, drums were assayed with this system. Certification included both debris and sludge waste types [10]. This instrument was placed into operation under the requirements of CH-WAC. Therefore, the system was required to have a daily quality control event, background event, a weekly interfering matrix test, and participate in an annual PDP test. The quality control events will not be addressed in this report. The daily operating procedure for WAGS Absolute was TPR-1654 [21].

LESSONS LEARNED

The lessons learned are broken into sections, the preliminary assay operation period and the production period. The preliminary assay operation is the time prior to September 29, 1997, the official startup date for production. The court approved milestone was met on September 30, 2003.

Preliminary Assay Operation (January, 1994 – September, 1997)

During this time period, the PAN assay system in combination with the SGRS system was the only radioassay method in service at SWEPP. The operating and analysis software was replaced with the Nuclear Quality Assurance (NQA) - 2 compliant software, object oriented software package – the SAS software operated on a 486 class PC. The major hardware additions were the SRC system in 1996 and the upgrades to the acquisition hardware in 1997, April – August. The PAN hardware upgrade in 1997 included a new neutron generator and pulse forming network, upgraded signal preamplifiers, and a digital link between preamps and gating module processors. Following the hardware upgrade a new baseline calibration was completed on the PAN assay system [22]. Based on the 1997 calibration, the PAN active mode range was from 0 g to 15 g and the passive mode had a range of 1 g to 160 g of weapons grade plutonium (WGPu).

After the new baseline calibration of the PAN assay system, a series of experiments were completed that assessed the impact of cosmic radiation to the PAN passive mode count rates. The PAN SRC system was allowed to run continuously to determine the frequency at which cosmic showers were detected. During that time period it was noted that cosmic showers occurred on a frequency of about once every 10 hours. The cosmic showers could be readily identifiable in the passive assay by a discrepancy between measured accidental coincidence count rate and the calculated accidental coincidence count rate. As a result, the SAS software included a provision to check the agreement between the measured and calculated accidental coincidence rates. Disagreement was a clear indication that a cosmic shower, or some other outside phenomenon, had occurred. Consequently, the occurrence of such an abnormality generated a simple error message to the PAN assay system operator. The corrective action, if such a message occurred, would be to repeat the assay.

As previously stated the PAN assay relied on default mass fractions for the plutonium isotopes. The original mass fractions from LANL were not substantiated by any references. Therefore, a study was initiated to arrive at the appropriate set of mass fractions to use for the plutonium isotopics in WGPu. This study examined over 2,000 shipping records from RFETS, which contained isotopic mass data. From this analysis a set of mass fractions were determined [23].

The first Performance Demonstration Program test, PDP Cycle 1, was completed in the summer of 1996. This PDP test showed that in order to meet the PDP criterion for precision, the PAN passive mode count time needed to be increased from 600 s to 1200 s on waste assays where the passive mode assay data could potentially be used as the reported data. However, this increase in count time was not required for waste forms where the active mode was the designated mode of choice; the active mode was the designated assay mode for sludge waste forms. This PDP cycle also further emphasized the need for total uncertainty tasks based on specific waste forms rather than broad based generic waste forms.

The original assumptions that were made in the early years were that the waste from RFETS was homogenous in nature, the only Am-241 present was grown in from the decay of Pu-241 in WGPu, and U-235 was not present. Assaying was originally done with the PAN assay system only. However it became apparent that the PAN assay results needed to be corrected for americium quantities above that which was accounted from the Pu-241 decay. Therefore a gamma system was added. Since the gamma system had multiple detectors it quickly showed the radioactivity was stratified in sludge waste as opposed to the original uniform distribution assumption that was used in the data analysis. Also, U-235 and U-238 were detected in the waste.

Due to the process time in a gamma system, it became apparent a second isotopic mass ratio (gamma) system was needed to improve productivity.

Production Assay Operation

There were several lessons learned during the 3100 m^3 Project that required changes to be made to the system. Some of the changes that occurred were due to the assaying of new waste types, while other changes occurred with the revisions of the WAC.

At the start of production, the only INEEL NDA system approved for radioassay was the PAN/Gamma assay system, consisting of the PAN system and the SGRS system. The primary waste form that was sent to WIPP prior to 2000 was graphite. The total measurement uncertainty (TMU) for graphite waste forms was completed in 1998 [24,25]. This waste form was chosen first because it was considered to be the most benign, as far as neutron absorption was concerned, and was expected to be the least challenging for the neutron based assay method, the PAN/Gamma technique. The first shipments to WIPP of graphite waste containers occurred in April of 1999. A significant number of containers of graphite had to be re-assayed or recalculated with SAS the software in Transuranic Reporting, Inventory, and Processing System (TRIPS) because the original TMU evaluation only included the assay results for the PAN's passive mode. Graphite containers that were re-assayed after the new requirements of the Waste Analysis Plan were incorporated into production, this occurred in the spring of 2000. The first lesson learned from this was if the original TMU had encompassed both the active and passive modes of the PAN/Gamma assay system recalculation efforts would not have been required. Secondly, recalculation as opposed to re-assay, was possible because the data collection procedures already met the requirements of the Waste Analysis Plan. Radioassay requirements at the INEEL exceeded requirements to assure data integrity.

The PAN/Gamma assay system's TMU evaluation was completed for aqueous sludge in 2000 [26]. Using the sampling/verification statistical approach for TMU analysis, radionuclide contents from radiochemical analysis of sample cores were correlated with the radioassay data collected by the PAN/Gamma assay system and were used to develop the TMU, bias and precision coefficients. The TMU limit was conservatively set, but this limit was later extended to be more reflective of the true NDA limit. The lesson learned was to fully implement the ranges in the beginning instead of modifying the certification limits later in time.

Beginning with SAS Version 3.0, the data from the PAN assay system was uploaded directly to TRIPS. Previously PAN assay results were uploaded to the RWMC Data Management System, which was later replaced by TRIPS. All radioassay data collected prior to the release of SAS Version 3 were required to be recalculated with the most current version of the SAS software before the data could be sent to WIPP. In 2000 all the data that had not been recalculated using SAS Version 3 and that were in the Radioassay Level 1 Data Validation queue were permanently rejected thereby requiring those containers to be re-assayed. The major lessons learned were to attempt to have the requirements for the radioassay systems, data acquisition, data reduction, data review, and database population as soon as possible, and make the systems flexible enough to evolve over time.

Evaluating the measured mass fraction data derived by the SGRS assay system was initiated to revisit the issue of default mass fractions derived from shipping records and the results of the new analysis are presented in EDF-1609 [23]. From a regulatory agency's prospective, the SGRS data should be considered more defensible because they were taken on a DOE Carlsbad Field Office (CBFO) certified instrument. The mass fractions determined in the SGRS data survey agreed to at least three significant figures with those determined from RFETS shipping records [27]. Because INEEL spent the time to determine the origin, handling, processes, packaging, etc. of its waste, the data collected was more accurate than the data provided by the waste generator.

For many of the waste assays there was a problem in measuring Pu-238 activity and Pu-240 activity leading up to the respective mass determinations. With respect to Pu-238 measurements, this isotope was present in very minute amounts in WGPu and in a large number of assays this isotope was not detected, or the measured activity had a very high uncertainty. Thus, the measured mass would be very uncertain at best. With respect to Pu-240, the decay of this nuclide produces only one viable gamma line at 160 keV. This gamma line is badly interfered with by gamma lines from the decay of other TRU radionuclides; hence, the resulting mass determination for Pu-240 was often suspect. Since Pu-239 activity was much easier to measure, the conclusion was that the plutonium isotope mass determinations should be based on Pu-239 using default mass fractions. The default mass fractions were in this case derived from measurements of waste drums in the INEEL inventory and were averaged over thousands of waste assays; hence, their uncertainties were much better than any single drum measurement. However, the SAS software checked the measured Pu-240/Pu-239 mass ratio and the Pu-241/Pu-239 mass ratio from the gamma system to assure that the plutonium in each individual drum measurement fit within the 95% confidence bounds of WGPu before it applied the default mass fractions. If a measured plutonium mass ratio datum was found to be outside those bounds, an error message was generated stating that the plutonium may not be weapons grade. Since americium and uranium were not covered, the measured mass of Am-241, U-235 and U-238 were reported in all cases. INEEL learned quickly that different regulatory viewpoints were often encountered in waste certification realm. Being able to identify the regulatory viewpoint of the auditor and defend the path chosen by the program was the best method for (re)certification of the NDA systems.

The SAS software stored the actual measured assay value in the SAS assay report file, even though it might be negative or less than the detection limits. Prior to the year 2000, negative or less than detection limit numbers were also uploaded to TRIPS. In the year 2000, negative or less than detection limit values were reported to TRIPS as a "-1", indicating a number that was technically not detected. These changes in the SAS to TRIPS data link did not change the way negative or less than detection limit values were reported to WIPP. TRIPS had always uploaded a "-1" to the WIPP Waste Information System indicating that the corresponding entry was looked for but not detected. The lessons learned were to clearly identify the sensitivity of the instruments and design software that could easily be modified for new WIPP requirements.

In December of 2000, the VAX workstation for the SGRS system was replaced with a PC with a Windows NT operating system. The SGRS acquisition and analysis software was replaced with SGAP. INEEL learned that all hardware and software has a finite lifetime. Maintenance and upgrades need to be planned for during the life of any project.

In December of 2000 the WAGS system was placed into operation to increase the throughput of the PAN/Gamma assay system. Because the WAGS system was similar to the SGRS system, the PAN assay system could then receive isotopic mass ratio data from either gamma system. The paperwork for the WAGS system was not sent to WIPP and this oversight resulted in suspension of shipments to WIPP. When shipments were reinstated to WIPP, the WAGS system was not allowed to support characterization until CBFO and Environmental Protection Agency had approved it. The WAGS system was approved for characterization in support of shipments to WIPP in August of 2001. The lesson learned was to verify the receipt of the paperwork by CBFO.

The following waste streams had TMU assessments completed using the general TMU evaluation method given in management control procedure (MCP), MCP-2990: mixed metals [28], raschig rings [29], glass [30], filters [31], and combustibles [32]. The following waste streams had TMU assessments completed using the general TMU evaluation methods given in MCP-2991: organic setups [33], plastics [34], solidified organics and special setups [35], and miscellaneous cemented waste [36].

Waste forms containing significant quantities of lead and other heavy metals was considered a problem for the PAN/Gamma assay method and also for the absolute gamma assay methods. A TMU evaluation for PAN/Gamma assays on lead waste forms was never completed. However, the TMU evaluation for SGRS Absolute covered lead containing waste forms where the heavy metal concentration was less than 38 kg [37]. Canberra Industries performed a supplemental TMU evaluation for WAGS Absolute for lead-containing wastes [38]. The limits of the scaling factors are: 1) plutonium 414/129 keV photopeak mass ratio between 2 and 10, inclusive; 2) matrix density between 0.2 g/cc and 1.0 g/cc, inclusive; and 3) TMU relative standard deviation greater than or equal to 15%. The lessons learned were to know the capabilities and limitation of each system.

EDF-2670 [39] was submitted to CBFO to extend the active mode calibration range from 15 g to 31 g of WGPu. With the approval of this document, approximately 800 drums were available for shipment to WIPP once the drums were reprocessed through data review processes. If full documented TMU limits and calibration ranges had been taken advantage of at the start of the certification process, this task would not have been needed.

The presence of U-233 in the INEEL TRU waste was a continuing issue. The SGRS (and the WAGS) gamma spectrum analysis would on infrequent occasions record a positive identification of U-233 activity based on the 317 keV gamma line from its decay. This particular gamma line is severely interfered with by the decay of Pu-239. To prevent an erroneous determination of the presence of U-233 the principal gamma energy for U-233 was changed from 317 keV to 291 keV. In addition, corroborating evidence required Bi-213, a decay daughter of U-233, be positively detected using the gamma peak at 440 keV. These modifications to the gamma assay analysis were implemented in the initial versions of SGAP and SAP.

The PAN assay system was potentially a single point for failure for the 3100 m^3 Project; therefore the decision was made to implement an absolute gamma assay system. A contract was awarded in 2001 to Canberra to convert the WAGS system to an absolute gamma system. Since the WAGS system had already been designed for this purpose this conversion did not require any physical changes to the WAGS system gamma enclosure however, a Ba-133 source and different detectors were installed. The conversion also required a change in the computer and the operating system, a change in the acquisition, control and analysis software. In addition to the hardware and software changes, a completely new calibration was required and completed. The WAGS system began operation as an absolute assay system in May of 2002.

As part of the INEEL Environmental Systems and Research Program (a research program separate from the 3100 m^3 Project), a method was developed whereby simple gamma spectrometers, like the SGRS system, could be used effectively for absolute assay. The 3100 m^3 Project office initiated implementation of the absolute assay analysis method on the SGRS system in 2001. The SGRS absolute software package was called SWEPP Absolute Assay Package (SAP). SGRS Absolute began operation in May of 2002. Initially, the SGRS absolute assay option was slated as a backup but it quickly was adopted as a primary absolute assay system. As a result, the PAN assay system was placed on standby and in August of 2002, the PAN assay system was deactivated.

During 2001 it became apparent that the PAN SRC system was overly sensitive to neutron multiplication in some waste drums where the plutonium mass was greater than 100 g. This was most evident in high-mass plutonium filter drums. Algorithms in the SAS software were being developed for high-mass plutonium drums to compensate for this inadequacy however, these algorithms were never completed prior to the end of the project. The alternative to assaying the drums with the PAN\Gamma assay system was to use SGRS Absolute, which was shown to be completely capable of assaying high-mass plutonium filter drums.

During the aqueous sludge campaign, an independent technical reviewer noticed that drums containing high amounts of U-235 relative to the Pu-239 concentration calculated the uranium and plutonium concentrations incorrectly. SAS Version 4.0 eliminated this problem, but was never installed on the PAN assay system because the 3100 m^3 Project could be successfully completed without the upgrade. The solution to the software upgrade was to assay those containers with SGRS Absolute.

In May 17, 2002, the new CH-WAC document was released by CBFO. The new CH-WAC required a lower limit of detection be determined for all the WIPP reportable quantities to be transferred to WIPP. If an assay measured value entry was less than its respective lower limit of detection, a message indicating such was to be placed in the data block in lieu of the measured value or zero. The new CH-WAC also removed the requirement of replicates and the

batch size restriction. The CH-WAC required weekly surrogate measurements to be performed. It also required reporting TMU at the one-sigma level rather than the two-sigma level, which was the case in earlier versions of the WAC. SAS Version 4.0 incorporated all new CH-WAC requirements. The absolute gamma systems were compliant with the new CH-WAC. Lessons learned were to implement the software immediately or find an alternative method for compliance.

EDF-2670, Revision 2 was released in June of 2002. This revision of the document extended the PAN active-mode range for solidified inorganic sludge wastes from 29 g to 47 g of plutonium fissile gram equivalent [40]. The lesson learned was that the full extent of TMU needs to be placed into operation once it was determined, being conservative increases operations costs at a later date.

The PAN/Gamma assay system participated in all the required PDP cycles and passed all tests with two exceptions, PDP Cycle 5 and PDP Cycle 6. Failure in PDP Cycle 5 was not the failure of the PAN/Gamma assay system but was due to a source distribution loading in PDP aqueous sludge drum that was outside the range found in sludge waste forms. Failure in PDP Cycle 6 also involved the aqueous sludge drum; in this case the plutonium loading was outside the calibration range of the PAN active mode. In both cases, corrective actions were sent to CBFO and provisional approval for continued operation was granted.

WAGS Absolute and SGRS Absolute participated in a PDP Cycle 8D before commencing assay operations. WAGS Absolute passed all tests and SRGS Absolute failed the bias test on the zero matrix drum. The failure was due to an erroneous drum mass entry in the SGRS assay parameter file. The zero matrix PDP measurement required a manual entry of the drum mass whereas under normal operating conditions the drum mass was entered automatically from a download of the data from TRIPS. Since a zero matrix was not a real waste form no corrective action was required.

In order to take advantage of those cases where data had been taken on the SGRS assay system under the mass ratio mode and certified the data for the absolute analysis mode, a provision was included in SAP, which allowed the previous data to be recalculated to produce absolute assay results without requiring re-assay. The decision was made to not implement this provision. Additionally when the SGRS Absolute software was placed in production, a decision to not implement any recalculation in TRIPS was made. This decision was the primary reason drums were not certified for shipment to WIPP by this instrument. The data validation process would often cause a drum to be reworked (i.e. such as reweighing the drum) after the radioassay had to be completed. The rework of the drum would not alter the data calculated/sent to WIPP but TRIPS would reflect newer information was available, and according to data validation procedures this would require the drum to be recalculated in TRIPS. The result was drums were permanently rejected because SGRS absolute was not WIPP certified to recalculate drums in TRIPS.

CONCLUSION

Over time, the RWMC Radioassay program adapted, modified, and expanded its methods and systems to meet changing requirements and needs over the entire lifespan of the project. The changes made and lesson learned were made to overcome limitations uncovered during testing and operations, to incorporate advances in hardware capability, to augment production throughput and reliability, to utilize improvements in algorithms, to include uncertainty determinations, and to improve data handling interfaces. As a result of a very active technical oversight team, general assumptions and broad statements were challenged and the experience gained should be a pattern to emulate in further NDA development and operations activities. It was found that each waste form and, in some cases an individual drum, represented a unique challenge in interpreting the assay data. As a result, the NDA hardwares/softwares/operating procedures were continuously updated and upgraded. The state of the assay systems and methodologies at the close of the 3100 m^3 Project reflects a long list of applied knowledge and experiences:

- There is no one assay technique that can be applied to all waste forms.
- The waste assay technique must be appropriate to the waste form and waste matrix.
- The knowledge of the waste form and packaging is absolutely necessary and should be as extensive as possible.
- The limitations of each assay technique must be well known.

- Calibration techniques, which are applicable for one technique, are not necessarily applicable to other techniques. As a result, calibration requirements need to be flexible enough to take advantage of advanced and different NDA methods.
- Calibration efforts must be rigorous and well documented.
- Throughput requirements should be well defined.
- Accuracy and precision goals should be well defined.
- Sensitivity goals should be well defined.
- Quality assurance methods should be well defined.
- Software and Program Documentation should be considered as "Living Documents".
- Automation of functions is the most desirable solution to "Special Cases".
- Advantageous to have well trained and experienced personnel.

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