

Characterization of Radioisotope Inventory In The Building 9204-3 Actinide Facility

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Introduction

An Extremely Brief History of a Beta-Calutron Facility (1940's – 1990's) - Building 9204-3, located within the Y-12 National Security Complex, is one of nine production facilities that were employed during the Manhattan Project for the enrichment of ^{235}U from natural uranium using electromagnetic isotope separation processes. After the end of World War II and the termination of the Manhattan Project, the nine facilities ceased uranium enrichment operations. Building 9204-3 (informally referred to as "Beta-3") subsequently was used in the 1950's through the late 1990's supporting the Oak Ridge National Laboratory (ORNL) Isotopes Program. The Isotope Program utilized the facility for two missions: Enrichment of specialty stable isotopes (Stable Isotope Program) and select actinide isotopes.

The portion of Beta-3 that was used for the enrichment of specialty actinide isotopes is referred to as the "Actinide Facility". It was comprised of dedicated calutrons and laboratories that were used to prepare feed stock/source material, and recover the product materials. The Actinide Facility conducted active isotope separation operations in the early 1960's up to the late 1970's. Actinide laboratory operations were terminated in the 1990's.

Fast Forward: Identifying Actinide Facility Radiological Inventory Issues - Although the Actinide Facility was no longer in operation, inventory records were still maintained for compliance with various requirements which included occupational radiological protection, facility nuclear safety, and asset management. These first inventory records tracked legacy feed stock/source materials and sealed sources. A critical look at the facility inventory records noted a lack of sufficient definition to estimate the amount of radioactive material present in the form of contamination and holdup. Legacy facility safety basis inventory requirements (dating back to ca. 1970's) did not specify any inventory tracking requirements nor any inventory limits, with the exception of several criticality safety and nuclear safeguards inventory limits.

Updating the Actinide Facility Radiological Inventory

Inventory Characterization Planning (2008) - In 2008, a team was convened for the purposes of providing recommendations for updating the Actinide Facility radiological inventory. The team reviewed the operational history of the Actinide Facility (e.g., procedures, log books, calutron operation summary data sheets, safety basis documents),

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facility design documents (e.g., schematics), interviewed personnel that previously worked at the Actinide Facility, and reviewed available radiological data (e.g. survey data sheets).

Late in 2008, the Characterization Planning Team issued a report which provided recommendations associated with four tasks specific to the successful implementation of a characterization plan:

Task 1 - Define the footprint of the Actinide Facility. Typical with legacy, non-operational facilities, the key to understanding the extent of the contamination inventory is to define all areas that supported the Actinide Facility. The footprint was determined by the team by defining (i) the specific calutrons used for actinide separation, (ii) the specific laboratories (and interconnected processes) that were utilized in the preparation and recovery of actinide materials, and lastly, (iii) all areas that provided support functions (such as storage of contaminated equipment).

Task 2 - Identify and prioritize materials, items, equipment, and processes that require characterization to ascertain radioactive material content. The team conducted a binning process to highlight those areas with a higher inventory potential from areas with lower inventory potential. This binning process allowed a characterization plan to appropriately allocate resources to the higher inventory potential areas, thereby obtaining the best possible estimate of the overall Actinide Facility inventory.

Task 3 - Identify appropriate characterization methods to be used to develop a radioactive material inventory. Using historical data, the team was able to define a spectrum of probable isotopes that would be present (mostly alpha emitters). The team recommended an approach that would utilize intrusive sampling (where possible) and an in-situ gamma spectrometer in order to obtain the best possible estimate of the radiological inventory.

Task 4 - Identify project tasks, materials, procedures, and personnel necessary to conduct a characterization sampling effort. Lastly, the planning team scoped the necessary resources that would be required to support the characterization plan. This scoping activity included a listing of necessary equipment repairs (particularly to the glove boxes), maintenance support, and also included a budgetary estimate (in terms of man-hours) that would need to be expended to complete the overall characterization activity. Additionally, the team identified significant safety and health issues that would need to be addressed to ensure a successful execution of the characterization plan.

Executing the Characterization Plan (2009 – 2011) - A Characterization Team was formed in 2009, consisting of facility support personnel (facility management, radiological protection, and maintenance organizations), project management, laboratory waste services, and nuclear facility safety.

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1. Constructing a Bounding Plutonium Isotopic Profile

In accordance with the characterization plan, the highest priority items were addressed first. This included areas specifically dedicated to the preparation, separation, and recovery of transuranium isotopes. The initial intrusive sampling activities focused on obtaining samples from the various vacuum systems. The vacuum systems were associated with systems and processes that involved the handling of volatilized materials. Log books from the 1960s, noted the need to frequently change the oil in certain support vacuum systems when the radioactive material concentration was too high. Initial scoping calculations indicated that the potential holdup in the vacuum system oils could be significant. After obtaining vacuum system oil samples, various laboratory analytical techniques were employed in order to define the isotopic content, including: (i) Gross Alpha [alpha spectrometry], (ii) ^{241}Am content [gamma spectrometry], (iii) Alpha Pulse [liquid scintillation], (iv) total activity [liquid scintillation], and (v) mass spectrometry.

Based on the cumulative analytical results, a bounding plutonium isotopic profile was constructed. The analysis reviewed the following plutonium profiles: (i) the isotopic content of the various vacuum system oils (from the intrusive sampling effort), and (ii) development of a composite plutonium isotopic profile based on historical data associated with recycle material. The analysis concluded the bounding profile was a 30-year decay corrected composite plutonium profile as developed from the recycle material historical data.

2. Incorporating the Bounding Plutonium Isotopic Profile Into In-Situ Gamma Spec Results

The analytical results from the vacuum system oil analysis was important for constructing a isotopic profile of the transuranium isotopes (mainly plutonium) that was not detectable in subsequent in-situ gamma spectrometry assessments of various equipment and areas of the Actinide Facility. In most cases, ^{241}Am tended to be the predominant gamma emitter detected via in-situ gamma spectrometry analysis. A review of the operational history determined the Actinide Facility engaged in a limited handling and processing of americium. Accordingly, the presence of ^{241}Am in the in-situ gamma spectrometry assessments was conservatively attributed as in-growth associated with the decay of ^{241}Pu . Since the Actinide Facility had ceased operations approximately 30 years prior to the start of the characterization plan, then the expected $^{241}\text{Pu}/^{241}\text{Am}$ activity ratio would be 9.54.

The derivation of the $^{241}\text{Pu}/^{241}\text{Am}$ ratio proved to be the key link between the in-situ gamma spectrometry data and a bounding transuranium profile (as derived from the laboratory analysis of the vacuum system oil). Based on the above, the bounding plutonium isotopic profile could be included in with the in-situ gamma spectrometry results as follows:

- Multiply the ^{241}Am activity content (from the in-situ gamma spectrometry data) by 9.54 to estimate the ^{241}Pu content.
- Use the activity ratio of ^{241}Pu to the other plutonium isotopes to derive the complete plutonium activity content of the equipment or area being assessed.

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3. Filling In The Blanks Via Decay Chain Analysis

Occasionally, other gamma emitters from the in-situ gamma spectrometry assessment would be reported that would not be associated with the plutonium decay chain. A review of the operational history of the Actinide Facility confirmed actinides other than plutonium were processed. A decay chain analysis was conducted to document the probable parent isotope responsible for the presence of the detected gamma emitter. Similar to the $^{241}\text{Am}/^{241}\text{Pu}$ methodology, a 30-year decay / in-growth was applied to determine the appropriate ratio's to be used to fill in the inventory. Examples included: (i) ^{214}Bi , ^{214}Pb (in-growth attributed to ^{230}Th decay), (ii) ^{212}Bi , ^{208}Tl (in-growth attributed to ^{232}U decay), and (iii) ^{228}Ac , ^{212}Bi , ^{208}Tl (in-growth attributed to ^{232}Th decay). Again, the inventory was adjusted to account for the probable presence of a parent that would not necessarily be detected via in-situ gamma spectrometry.

4. Modeling Activity Content With Microshield

Several items associated with the Actinide Facility were of complex geometry or involved complex attenuation / shielding situations. The geometry simplifications incurred using rudimentary models associated with the in-situ gamma spectrometer would lead to extremely conservative (very high) inventory estimates. On several items, a refined inventory assessment was made by making detailed gamma dose readings around the item and inputting the data into a Microshield® software application. The predominant gamma emitter used in the analysis was ^{241}Am (based on the gamma spec analysis). The Microshield model provided a refined estimate of the ^{241}Am inventory. The refined ^{241}Am inventory was then used to derive the plutonium content (using the bounding plutonium profile).

5. Completion of Phase 1 of the Characterization Plan & The Initiation of Phase 2

By the end of 2009, the first phase of the characterization plan was completed. The highest priority items had been assessed and inventory estimates were documented in detailed calculation reports. Using the techniques mastered from the first phase of the effort, Phase 2 was initiated.

Phase 2 involved an assessment of the lower priority items as identified in the 2008 characterization planning report. The assessment included extensive use of the in-situ gamma spectrometer for specific items and small areas. For inaccessible items (e.g., obstructed contaminated equipment) and larger areas/items (e.g., rooms, ventilation systems), radiological survey data sheets were used to estimate the radiological inventory.

Generally, the items assessed in Phase 2 were low in inventory as anticipated by the Characterization Planning Report. However, a storage area was noted to have contaminated items with significant gamma dose rates. A review of the operational usage of the high dose rate items indicated a high probability of direct contact with plutonium recovered for processing. As noted similarly in other cases, the predominant gamma emitter was ^{241}Am (which is consistent with items that would have been in contact with plutonium material).

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Actions Taken After Completion of the Characterization Plan (2011 - 2012) - With the completion of the second phase of the characterization plan, the accumulated data now represents a complete estimate of the facility inventory. Armed with the entire inventory data set, a facility hazard categorization could be documented.

1. Documenting the Safety Basis

The initial hazard categorization determined the facility inventory exceeded Hazard Category 3 thresholds, as determined by a sum of fractions (SOF) assessment. Since the fissionable material inventory was below the Hazard Category 2 fissionable mass thresholds, a final hazard categorization was conducted in accordance with NSTP-2002-2. The bounding hazard for Beta-3 was determined to be a seismic event (complete facility collapse with an impact type release of material), followed by a subsequent fire (release of radiological materials via exposure to thermal stress). The bounding event was determined to have a modified SOF less than 1.0, thereby resulting in the facility having a final designation as below Hazard Category 3 (e.g., a Radiological, By Analysis facility).

2. Reduction of Risk Accomplished During the Characterization Plan

The Characterization Team utilized the knowledge they accrued during the project to take actions to mitigate the risks associated with the presence of radiological materials in the facility. The team was able to change out old, deteriorated gloves with new gloves in the various glove boxes and enclosures throughout the Actinide Facility. Additionally, updated ventilation instrumentation was equipped to the glove boxes, and HEPA filters on the glove boxes and enclosures were changed to improve the integrity of the ventilation flow through these contaminated systems. Glove boxes and miscellaneous areas were cleaned out of unneeded items (thereby reducing the inventory associated with these items) and several high inventory items were removed from the facility in order to minimize the final facility inventory.

Summary

Through a dedicated and deliberate process that spanned several years, ORNL was able to accurately determine the radiological material inventory associated with holdup and contamination that had previously been uncharacterized. The process of identifying the location and amount of inventory allowed ORNL to manage the inventory so as to reduce risks. The final hazard categorization is now based on an inventory that reflects the actual conditions of the facility.

Acknowledgements

The author of this paper would like to extend his sincerest appreciation to members of the Beta-3 Characterization Planning Team, the Beta-3 Characterization Project Team, the Beta-3 Facility Management, and the members of the ORNL Nuclear & Radiological Protection Division (NRPD) for their dedication and professionalism in working together to compliantly achieve all characterization project goals.