RH-TRU WASTE CHARACTERIZATION BY ACCEPTABLE KNOWLEDGE AT THE IDAHO NATIONAL ENGINEERING AND ENVIRONMENTAL LABORATORY

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ABSTRACT

Idaho National Engineering and Environmental Laboratory (INEEL) is conducting an effort to characterize approximately 620 drums of remote-handled (RH-) transuranic (TRU) waste currently in its inventory that were generated at the Argonne National Laboratory-East (ANL-E) Alpha Gamma Hot Cell Facility (AGHCF) between 1971 and 1995. The waste was generated at the AGHCF during the destructive examination of irradiated and unirradiated fuel pins, targets, and other materials from reactor programs at ANL-West (ANL-W) and other Department of Energy (DOE) reactors. In support of this effort, Shaw Environmental and Infrastructure (formerly IT Corporation) developed an acceptable knowledge (AK) collection and management program based on existing contact-handled (CH)-TRU waste program requirements and proposed RH-TRU waste program requirements in effect in July 2001. Consistent with Attachments B-B6 of the Waste Isolation Pilot Plant (WIPP) Hazardous Waste Facility Permit (HWFP) and the proposed Class 3 permit modification (Attachment R [RH-WAP] of this permit), the draft AK Summary Report prepared under the AK procedure describes the waste generating process and includes determinations in the following areas based on AK: physical form (currently identified at the Waste Matrix Code level); waste stream delineation; applicability of hazardous waste numbers for hazardous waste constituents; and prohibited items. In addition, the procedure requires and the draft summary report contains information supporting determinations in the areas of defense relationship and radiological characterization.

In addition, a separate radiological characterization report, "Radiological Acceptable Knowledge Report for Drums 728 through 737," (INEEL/EXT-02-00527) was also prepared and finalized in June 2002. The report provides detailed radiological information for one ten-drum batch using AK documentation. For this effort, hundreds of AGHCF and INEEL records were investigated, including data on pre-irradiation compositions, reactor operational history, post-irradiation composition calculations and

ORIGEN-based modeling results, accountability tracking forms (such as Loss-Gain and Material Receipt forms), and AGHCF records (A/G books) maintained for each piece of each fuel pin or other item examined. INEEL is proposing a radiological characterization method based on AK for RH-TRU waste generated at ANL-E. Use of this information may reduce unnecessary radiation exposure to workers while meeting all regulatory requirements for transportation and disposal of RH-TRU waste.

INTRODUCTION

Shaw Environmental & Infrastructure (Shaw E&I) compiled, and is currently reviewing, AK documentation for INEEL regarding approximately 620 drums of RH-TRU waste that is stored at the facility’s Radioactive Waste Management Complex in Idaho. The RH-TRU waste investigated was generated at ANL-E during the operation of the AGHCF from 1971-1995.

The RH-TRU waste was generated in the AGHCF from the destructive examination of irradiated and unirradiated fuel pins, reactor structural materials, and targets, as well as maintenance and repair operations. The purpose of this paper is to discuss the program and AK reports developed for this effort and future work that is currently anticipated. In support of the AK effort, a full program was developed comprising nine procedures and plans that covered AK collections and determinations, quality assurance (QA), training and qualification, records management, and document control. Upon completion of the program development phase of work, the AK source documents were collected and a draft AK summary report was prepared that documented overall descriptions of the RH-TRU waste generated. A supplemental report was also prepared to provide detailed radiological characterization of a ten-drum subset of the population. Future work for the project involves compliance with final WIPP RH program requirements and providing detailed radiological characterization of the remaining inventory.

Site Description

The University of Chicago has operated ANL-E since 1941 for the DOE and its predecessor agencies (Atomic Energy Commission [AEC], Energy and Resource Development Agency [ERDA]). ANL-E was established as a result of early experiments conducted at the University of Chicago to produce plutonium for nuclear weapons and was officially founded in 1946 as the first national laboratory. ANL-E has been actively involved in the development of nuclear reactors and associated systems, fuel elements, materials, and components for both civilian and defense programs.

RH-TRU waste was generated from the destructive examinations and maintenance operations undertaken at the AGHCF. The AGHCF is located in Building 212 (F-wing) on the ANL-E site. A diagram of the AGHCF layout is provided in Figure 1. The facility includes the actual hot cell, comprised of ten work stations located in Rooms F-131 through F-136, and support areas including offices, showers, and dressing rooms.

The AGHCF is unique in that it is the only hot cell in the United States with the capabilities to perform certain types of destructive testing in conjunction with other examination and testing methods in the same remotely controlled environment. It is constructed of a single hot cell

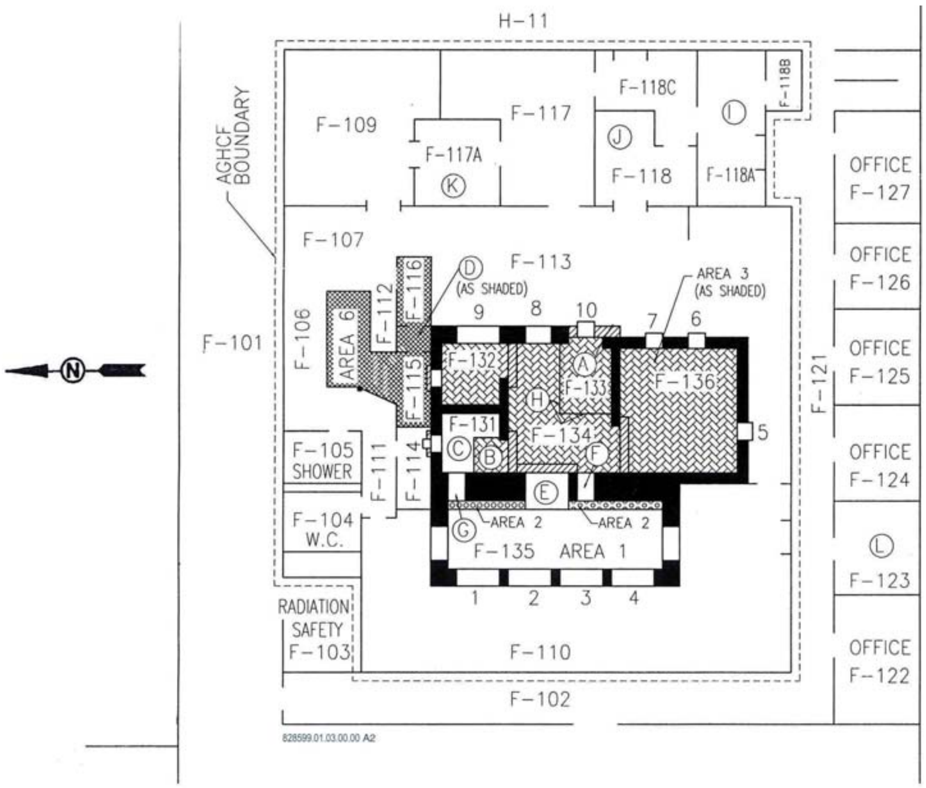

Fig.1.
Alpha Gamma Hot Cell Facility

(under a nitrogen atmosphere) divided into three main areas subdivided into ten workstations. The following is a partial listing of the examinations performed in the hot cell:

Nondestructive Examinations

Initial identification, visual examination, and handling
Capsule disassembly
Macrophotography of irradiated fuel elements
Bow measurement
Diametral profilometry
Diameter measurements with micrometers
Eddy current inspection
Fuel element length measurement
Irradiated fuel element balance point
Weighing procedure
Irradiated fuel element helium leak test
Autoradiography
X-ray procedure and gamma scanning

Destructive Examinations

Fuel element fission gas sampling and H-3 gas collection
Plenum volume determination and gas analysis
Fuel element sectioning
Mounting metallographic specimens
Preparation of mounted metallographic specimens (includes grinding of samples)
Metallography
Microphotography
Microhardness measurement
Determining specimen density by immersion
Replication for microscopy

These operations generated a fine fuel particulate of swarf that adhered to the tools and equipment required to support hot cell operations, thus contaminating them with TRU elements. These tools (e.g., cutting wheels, grinding papers, glassware, light bulbs, and rags used for cleaning) were sent to INEEL as RH-TRU waste. Distinct pieces of fuel elements, cladding, irradiated structural material, and recovered swarf and fines were not part of RH-TRU waste sent to the INEEL (they were considered "high level" waste or spent nuclear fuel and were placed in one gallon cans and stored in the AGHCF or returned to the originator of the fuel).

PROGRAM DEVELOPMENT

The AK collection and management program developed and implemented for INEEL included nine different procedures and plans developed over a two-month period. The program was designed to meet contact-handled (CH) TRU waste program requirements current as of July 2001 when the project was initiated and RH-TRU waste characterization requirements proposed at that time.

The AK Information Collection Procedure and Quality Program Plan (QPP), along with other implementing procedures, were used to perform work for this program. Waste characterization using AK involves the compilation and evaluation of information concerning the waste generating processes or activities at a site. AK information may include previous testing data, waste generating procedures, chemical inputs to the processes, time period in which the processes took place, the descriptions of the facilities involved, types of waste generated, and hazardous waste determinations made to satisfy host state requirements.

Under the existing CH-TRU waste program for waste to be disposed at WIPP, AK is required to be used in TRU waste characterization activities and this program to:

- Delineate waste streams
- Assess whether heterogeneous debris wastes exhibit a toxicity characteristic
- Assess whether wastes are listed.

Under the proposed RH-TRU waste program, AK may also be allowed to support determination of the physical waste form, the absence of prohibited items, and the radiological properties of the waste.

As the WIPP remote-handled RH-TRU waste characterization program has not yet been finalized, this AK Information Collection Procedure conservatively applied the CH-TRU Waste Analysis Plan (WAP) requirements to an INEEL’s RH-TRU waste inventory (1). In addition, proposed RH-TRU waste characterization requirements at that time contained in the draft RH WAP and Waste Characterization Program Implementation Plan (WCPIP) were incorporated. The procedure implements the CH-WAP requirements for the assembly, compilation, evaluation, and reporting of AK documentation, as well as the requirements for resolving discrepancies in AK documentation.

The AK Information Collection Procedure details the types of AK source documents and the methods for collecting them. AK source documents may include communications (interviews with generator personnel, internal memos, letters, and e-mail), published and internal documents, site/waste generator procedures, and data collections (analytical data reports and databases), and any other pertinent types of documentation. The methodology for collecting AK information developed for this program is similar to that used for AK information collection by the WIPP Central Characterization Program at the Nevada Test Site, Los Alamos National Laboratory, and the Savannah River Site, including a checklist of required information and summarizing source documentation.

The QPP and supporting implementing procedures were developed to meet the requirements of the WIPP Quality Assurance Program Document (QAPD), the governing quality assurance (QA) document for the TRU waste disposal program (2). These quality assurance requirements include the proper handling/control of records and documents, AK information collection personnel training requirements, and other supporting QA functions. The program was judged to meet the requirements through independent review and auditing.

AK SUMMARY REPORT

The initial phase of the AK collection resulted in the preparation of a draft AK summary report covering two waste streams, IN-W259-C (combustible waste) and IN-W259-NC (non-combustible waste). This summary report presents data collected from more than 250 different sources relating to historical ANL-E and ANL-W operations, waste generating processes, and waste management practices. The objectives of the report are to summarize the following major essential elements: defense determination, waste generating processes and waste management procedures, area and building of generation, physical form, chemical content and hazardous waste numbers, prohibited items, and radiological characterization.

AGHCF Defense-Related Activities

ANL-E conducted a variety of different types of nuclear reactor research in its 55-year history, including the design and building of several reactor prototypes on its campus in Illinois and at ANL-W. During the 39 years that the AGHCF has been in operation, many defense-related programs have been conducted in the facility. In most cases, the AK information compiled for this effort shows that consolidated batches used to track groups of waste containers included residue from several different fuel elements and often from different programs, at least some of which were defense-related (i.e. Experimental Breeder Reactor [EBR]-II and the Integral Fast Reactor [IFR], located at ANL-W). Based on review of fuel pins listed in the consolidation batch, review of historical documents, and a review of consolidation batch data, approximately 92% of the material examined in the hot cell came from ANL-W reactors. Non-defense related programs known to have submitted samples to the AGHCF include Three Mile Island and Chicago Pile-5. Wastes from defense related and non-defense related programs are commingled throughout the inventory of waste stored at INEEL (3).

In addition to the apparent commingling of material from different sources, AGHCF personnel estimated that 25% of the swarf, or cutting residue from the cutting of fuel rods, was left in the hot cell as adventitious dust even after typical clean-up efforts, which were conducted infrequently. A percentage of the total swarf was assumed to be on the cell's fixed surfaces and items such as tools and light bulbs that eventually were packaged as waste. As a result, there was some commingling of material among consolidated batches, in addition to the commingling of material within each batch. As a result, RH-TRU waste drums generated from 1971 to 1995 at the AGCHF at ANL-E contained commingled defense and non-defense waste that could not be physically separated, which qualifies it for disposal at WIPP under the terms of the Nordhaus memorandum. However, a formal defense determination will be made upon completion of the AK Summary Report.

Waste Generating Process

The types of investigations performed in the AGHCF have been described previously. Examination worksheets or work plans outlining the tests that were or were to be performed on each samples were collected for this effort. Most of the destructive examinations were performed in Work Stations 1-4 of the AGHCF hot cell, while waste was packaged at Work Station 5. Waste was also generated by routine equipment maintenance and repairs, including replacement of saw blades, broken glassware, gloves, and light bulbs, to less routine tasks such as replacement or repair of experimental apparatus and electrical motors. Waste was usually collected in cans at the point of generation and then moved to the packaging area at Work Station 5 for storage, where it was segregated and packaged into 7.5-gallon cans and removed from the hot cell for packaging into 30-gallon drums.

Documentation required for each can and drum changed over the 1971-1995 time period of waste generation, but began with the "Intermediate Gamma-Level TRU Packaging" form on which generators recorded inner can numbers, drum and seal number, packaging date, and smearable and contact activities, in addition to accountability documentation that described accountable radiological contents. Documentation requirements increased over time to include

Pu-239 and U-235 grams, removable contamination, and, by 1985, presence/absence of liquids, pyrophorics, pressurized vessels, corrosives, prohibited items, and drum contents.

Physical Form Determination

From the compiled AK documentation, the physical form of the waste streams was determined. The RH-TRU AGHCF waste stored at INEEL had been segregated into combustible and non-combustible streams. The requirement to segregate combustible and non-combustible TRU waste originated from Atomic Energy Commission (AEC) Manual, Chapter 0511, which defines combustible waste as "Organic material capable of being burned," except for noncombustible waste packaged in plastic (1973) (4). The following definitions were developed and repeated in AGHCF Safety Analysis Reports (5, 6):

- Non-combustible debris: Ferrous metals, including carbon and stainless steel and cast iron; nonferrous metals, including aluminum, brass, bronze, copper, lead, and tin; glass bottles, tubing, beakers, and plates; ceramic firebrick, porcelain (insulators), quartz, Vycor, and boron nitride; and "passivated chemicals" absorbed in clay.

- Combustible debris: "Neoprene gloves and O-rings, polyethylene and polypropylene bottles; plastic tubing (including poly vinyl chloride [PVC], polyethylene, rubber, and styrene butadiene); PVC, polyurethane, and polyethylene bagging pouches; silicone and Teflon O-rings; paper products; cotton and synthetic rags; polyethylene and PVC sheeting; wood products (including masonite); neoprene, koroseal, and rubber gaskets; and a variety of plastics and cellulosics."

Consistent with CH-TRU waste program requirements, waste physical form has been described in terms of waste matrix codes in the AK Summary Report. From 1990 forward, generators were required to estimate the volume percent of each type of waste on packaging inventory sheets. Descriptions of waste contents provided for waste generated before 1990, videotapes of packaging available from 1993 forward, and other information from disposal requisitions was used to delineate waste drums into the combustible and non-combustible streams. Because of the implemented and enforced segregation that was maintained throughout the period of waste generation, each waste stream is estimated to contain greater than 80% by volume of either organic or inorganic waste constituents. The following waste matrix codes are applied: for waste stream IN-W259-C, S5300, Organic Debris; and for waste stream IN-W259-NC, S5110, Metal Debris.

Chemical Usage and WIPP Hazardous Waste Number Determination

Numerous references were investigated to describe chemical use in the ANL-E AGHCF, including correspondence, safety analysis reports, procedures for activities conducted within the hot cell, Material Safety Data Sheets, generator interviews, and data recorded by AGHCF technicians on can inventory sheets and disposal requisitions. Chemicals used in the AGHCF hot cell included acid etchants such as sulfuric, hydrochloric, chromic, phosphoric, nitric, citric, acetic, and oxalic acids, silver nitrate etch, and various solvents such as toluene and 1,1,1-trichloroethane used for cleaning manipulator arms. Other materials found to be present in the

RH-TRU waste drums that could cause waste to be characterized as hazardous (mixed) if present in excess of the regulated quantities include:

- Lead in manipulator weights, as scrap, cerrobase epoxy, solder, and batteries
- Mercury in thermometers and mercury vapor lamps
- Silver in silver nitrate etch, epoxy, and solder
- Sodium (ignitable) reacted with alcohol and evaporated to dryness

ANL-E consistently managed the AGHCF RH-TRU waste containers under investigation as RH-TRU nonhazardous waste. Technicians completing waste disposal requisitions did not identify the presence of RCRA hazardous wastes or mixed waste constituents from the time, beginning in 1984, that these forms had space available to record such information. Shaw E&I personnel are reviewing AK documentation to determine if any applicable hazardous waste numbers need to be assigned for the disposal of this waste at WIPP. As required by the CH-WAP and proposed RH-WAP, INEEL will conservatively apply applicable hazardous waste numbers (if any) to the RH-TRU waste for disposal of the waste at WIPP.

Prohibited Items

Throughout the period of waste generation under investigation, liquids, compressed gases (including aerosol cans), pyrophorics, explosives, and other incompatible or "potentially reactive chemicals" were prohibited from the waste stream based on compiled AK. Liquids have been prohibited in RH-TRU waste since and continued to be prohibited in the 1986 TRU-RH Waste Certification Plan for Waste Management Operations and later revisions (7, 8). From at least 1977, INEEL Waste Acceptance Criteria (WAC) also prohibited liquids, pyrophorics, and potentially reactive chemicals.

No polychlorinated biphenyl (PCB) contamination is anticipated to be present in this waste stream. All electrical equipment that might have contained PCBs (such as ballasts and capacitors or transformers) was located outside of the AGHCF on the second floor of the wing for ease of maintenance. The only oil used inside of the Hot Cell was in a pressure relief reservoir in the facility exhaust system, but this fluid was never been changed out and, thus, would not have entered the RH-TRU waste stream.

Liquids are known not to be present in waste containers, due to standard practices to evaporate or absorb all liquids in vermiculite or pelletized clay of all liquids, as well as the fact that no more than three liters of hydrogenous liquid were allowed in Areas I or III of the hot cell.

RADIOLOGICAL CHARACTERIZATION

The majority of fuel pins (92%) examined in the AGHCF were irradiated at the EBR-II reactor at ANL-W. Excellent pre- and post-irradiation information, including run history, is available. For the fuel pins irradiated at ANL-W, calculations to estimate post-irradiated compositions were performed using BURNOUT, a software code that calculated fuel depletion in the EBR-II core. The information from the BURNOUT code was used to track the depletion and generation of fissile material in support of reactor research and operations. The results were also used to complete safegurads and security forms (Form 741s) when fuel elements were transferred from ANL-E to ANL-W for examination.

For ANL-E fissile materials tracking, all nuclear material that was transferred into (or out of) the AGHCF required Special Materials (SPM) documentation in the form of a "Receipt – SS Materials" form (Form SPM-24), which relied on information supplied on the 741s form supplied by ANL-W. The fissile material lost to RH-TRU waste was calculated for each batch and recorded on the "Loss or Gain – SS Materials" form (Form SPM-23). The information from the SPM-23 was provided to Waste Management Operations (WMO) on the then current version of the waste requisition forms. At that point, the RH-TRU drums were dispositioned to WMO and eventually shipped to INEEL.

Also, each accountable item in the AGHCF was assigned an Alpha/Gamma (A/G) Number upon arrival. These numbers were based on a combination of numbers and a single letter used for sample tracking. Items that were from the same program and arrived together, could use the same leading serial number but with a unique trailing letter. Consequently, a three-ring binder was then created and labeled with the A/G number and termed an "A/G book." The A/G books contain information regarding the item's composition and origin as well as detailed accounts of the examinations performed.

The necessary examinations were then performed on the specimens and recorded in the A/G books and on examination-specific sheets, where applicable (such as the Grinding Loss Record). ANL-E also utilized a database, called the "Fissile Inventory Management System Database" to track accountable quantities of fissile materials. Another database maintained by the AGHCF, LANARK (formerly SIMS), correlated batch numbers entering the AGHCF to A/G Nos. for particular samples, material, type, and description.

These records and databases were used to develop the list of fuel pins or items examined that contributed to a particular waste packaging campaign. This process is referred to as batch consolidation.

Batch Consolidation

According to the AGHCF Operations Manual, personnel estimated the amount of fissile material in a batch of drums by assigning a cut-off date for the last waste to be moved from Area I (where most destructive examinations occurred) to Area III (Work Station 5) and determining the number of fuel pins sectioned since the last cut-off date. The pins were included in the consolidation batch and the total fissile content for the batch of drums was determined.

The AGHCF waste generators completed Form SPM-23, "Loss or Gain—SS Materials," to quantify how much material from each fuel element was "lost" and how much was "gained" by the TRU waste. These calculations were developed in the mid-1970s to quantify Pu-239 and U-235 to meet nuclear accountability requirements. More specifically, hot cell waste was allocated amounts of fissile material generated by the activities taking place in the hot cell during the period of waste generation, including sectioning, cutting, grinding, and mounting of specimens. Ideally, all of the source fuel elements that lost material between the dates noted on the AGHCF Records Chief Technician's notes and corresponding Loss/Gain forms would be accounted for under a new "consolidated batch number" assigned to the waste drums. All of the material lost from the elements would be added to (or "gained by") the new consolidated batch number assigned to the waste. Radionuclide contribution was not calculated for items examined in the hot cell that did not contain fissile material.

The assumptions used to estimate fissile material loss from an average fuel element and based on operational history were as follows:

1. Average fuel element diameter—0.2 in.

2. Average fuel element length—13 in.

3. Average five mounts or ten cuts per pin, four mounts transverse and one mount longitudinal in orientation

4. Saw blade width (thickness)—0.04 in. (average width of blades 0.032 and 0.050 in., with 75% of the blades having the 0.032-in. thickness)

5. Transverse mount removal of 0.030-in. material from the fuel element during grinding; longitudinal mount removal of 0.75 in., half of which is fuel (the other half is insulation and is not fissile material).

Volume of Material Lost from 4 Transverse Mounts:

$$\pi r^2 \left[(Width\_lost\_from\_cuts) + (Width\_removed\_for\_samples) \right] =$$
$$\pi r^2 \left[(8 * .04) + (4 * .03) \right] = 0.44 * \pi r^2.$$

Volume of Material Lost from 1 Longitudinal Mount

$$\pi r^2 \left[(Width\_lost\_from\_cuts) + (Amount\_removed\_for\_samples) \right] =$$
$$\pi r^2 \left[(2 * .04) + (0.375) \right] = 0.455 * \pi r^2.$$

Total Lost from Element = (0.44 + 0.455) πr^2 = 0.895 πr^2

Total Volume of Element = Assume 13" element, so that Volume = 13 πr^2

% of Element Lost: 0.895/13 = 6.88%

The swarf generated from the 6.88% of the element lost during hot cell operations was divided among the recoverable high-level sweeps, adventitious dust remaining in the hot cell, and the RH-TRU waste.

Based on operational experience operators were able to recover 75% of fuel fines and these fines were collected and stored in hot cell as high level waste. The remaining 25% of the swarf either adhered to the fixed surfaces in hot cell or to the equipment and tools used in the cell's operations. The 25% of swarf amounts to 1.72% of element (0.25x6.88%). Considering the effectiveness of the swarf recovery/collection process, from cutting and grinding operations, the AGHCF's practice was to use 1.5% of the fuel element as an estimate of what is in the waste (9), with the rest of the swarf remaining in the hot cell. The information described above was used to form a basis of the fissile radionuclides including Pu-239 and U-235. Cs-137 quantities were determined based on measured dose rates.

As mentioned previously, in addition to the AK report for the two subject waste streams, a separate report was compiled to present the radiological characterization for one batch of ten drums of RH-TRU waste from the AGHCF generated from August 28, 1990 and July 17, 1991. For this task, data was compiled from both ANL-E and ANL-W regarding the exact pins examined, examinations performed, pre- and post-irradiation compositions, and reactor run histories. INEEL used this information to determine a more precise radionuclide contribution (than the assumed 1.5% of fuel pin described above). The information was compiled in a report (10) and is being used by INEEL personnel in demonstrating the radioactive waste characterization methodology using AK to meet the WIPP transportation and disposal requirements. This methodology utilizes the QA provisions of 40CFR 194.22(b) to qualify AK-based characterization by peer review. Results of the work being performed are the subject of a poster session, which is also being presented at Waste Management '03.

FUTURE WORK

Future work for this program will entail the evaluation of AK documentation and any supplemental information to determine if any hazardous waste numbers need to be applied to the waste streams for disposal at WIPP, the completion of the AK Summary Report, and the radiological characterization of the remaining inventory of AGHCF waste stored at INEEL. In addition it will be necessary to evaluate the INEEL program and bring it into compliance with the approved RH program requirements, WCPIP and the HWFP (i.e. RH WAP).

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