PLANNING FOR THE MANAGEMENT AND DISPOSITION OF NEWLY GENERATED TRU WASTE FROM REDC

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ABSTRACT

This paper describes the waste characteristics of newly generated (NG) transuranic (TRU) waste from the Radiochemical Engineering and Development Center (REDC) at the Oak Ridge National Laboratory and the basic certification structure that will be proposed by the University of Tennessee – Battelle (UT-B) and Bechtel Jacobs Company LLC (BJC) to the Waste Isolation Pilot Plant (WIPP) for this waste stream. The characterization approach uses information derived from the active production operations as acceptable knowledge (AK) for the REDC TRU waste. The characterization approach includes smear data taken from processing and waste staging hot cells, as well as, analytical data on product and liquid waste streams going to liquid waste disposal. BJC and UT-B are currently developing the elements of a WIPP-compliant program with a plan to be certified by the WIPP for shipment of newly generated TRU in the next few years. The current activities include developing interface plans, program documents, and waste stream specific procedures.

THE REDC

The Radiochemical Engineering and Development Center (REDC) began operation in 1966 as a reactor target processing and isotope recovery/production facility (1). The REDC continues in this role today and is currently supporting the Transuranium Element Processing Program, the Mark-42 Processing Program, and the Californium-252 Industrial Sales/Loan Program. All three of these programs operate concurrently or alternately in Buildings 7920 and 7930. Building 7920 and Building 7930 are shown in Figure 1. These programs are co-sponsored by the Department of Energy (DOE) Office of Nuclear Materials Production, Defense Programs, with approximately half of facility funding provided by the DOE Office of Defense Programs.

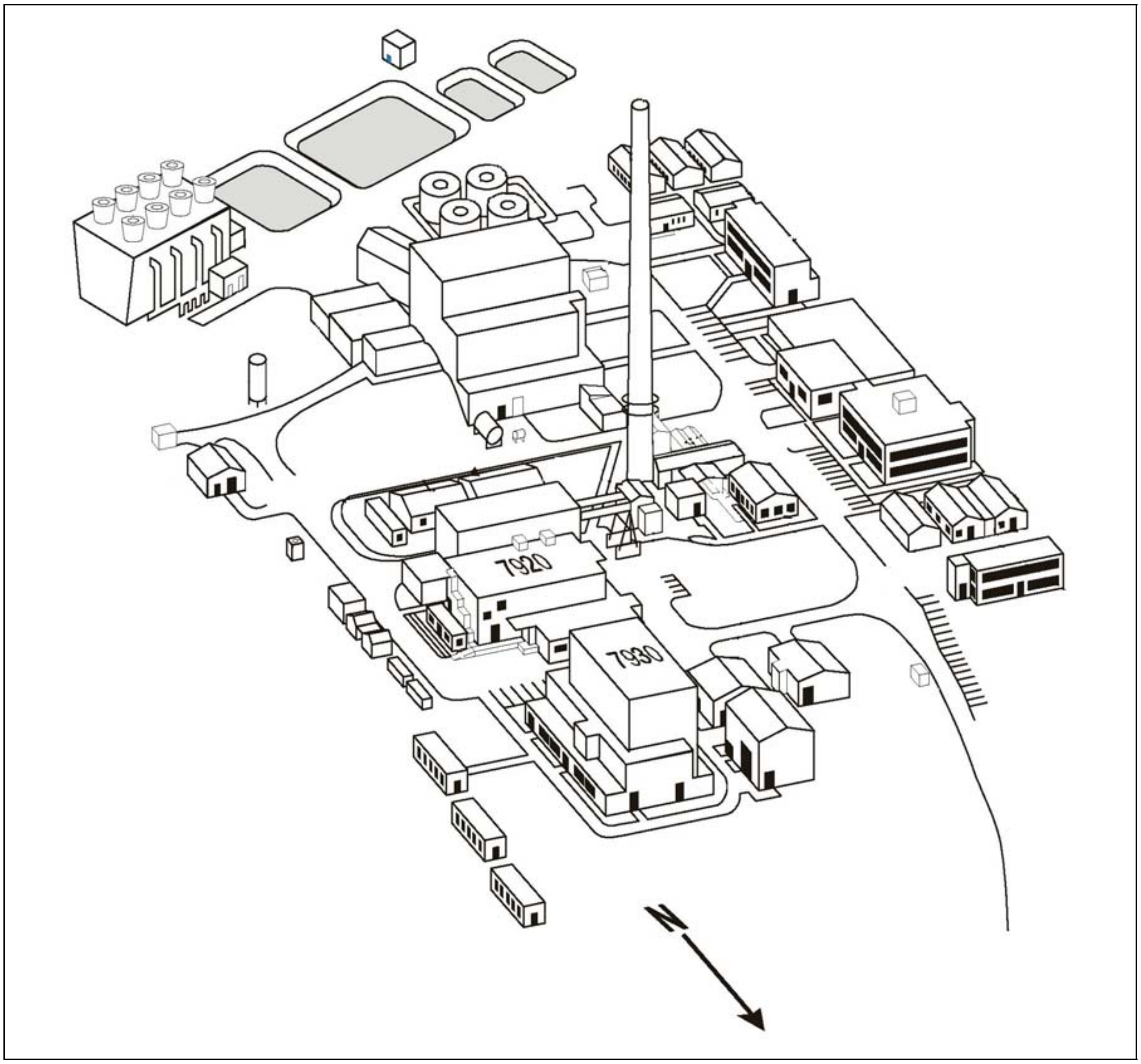

Fig. 1. Radiochemical Engineering Development Center (REDC), Buildings 7920 and 7930

The Transuranium Element Processing Program supplies critical isotopes for research activities to both the DOE weapons laboratories and the multi-program research, design, and development (RD&D) laboratories. Isotopes produced under this program include Am-243, Bk-249, Cf-249, Cf-252, Cm-244, Cm-248, Es-253, Es-254, Fm-255, and Fm 257 as well as various fission products. The Mark-42 Program recovers TRU elements, including Am-243, Cm-244, and Pu-242, from reactor targets.

The Californium-252 Industrial Sales/Loan Program provides Cf-252 neutron sources to more than 70 institutions (2). These institutions consist of DOE or integrated contractors, U.S. Government agencies, including military installations, educational and medical institutions, and private research organizations. The uses of these sources include medical research or treatment, classroom instruction or demonstration, and R&D or industrial applications (e.g., reactor startup, radiography, neutron activation analysis, explosives detection). Specific defense/military uses of the Cf-252 neutron sources include:

- Nondestructive inspection of explosive fill in detonators at Mound,
- Neutron activation analysis at the Savannah River Site (SRS),
- Assay of high-level glass at the Hanford Site, Nondestructive assay systems for fissile and TRU wastes at Los Alamos National Laboratory (LANL) and the Idaho National Engineering and Environmental Laboratory (INNEL),
- Neutron radiography to locate corrosion on military fighter aircraft at McClellan Air Force Base, and,
- Substitute reactor at the Naval Ocean Systems Center.

Generator/storage sites must also demonstrate that their waste is not spent nuclear fuel or high-level waste as defined in the *Waste Isolation Pilot Plant Land Withdrawal Act*. Spent nuclear fuel is fuel that has been removed from a reactor but has not undergone reprocessing. The hot cell waste consists of miscellaneous debris from isotope recovery and purification, not spent nuclear fuel. Although irradiated actinide targets are processed for isotope recovery, they do not end up in the waste. Therefore, TRU waste generated from the isotope recovery/production operations at the Building 7920 and 7930 hot cells is eligible for disposal at WIPP. It is generated in whole or in part from the following defense activities: naval reactors development (as a substitute naval reactor), verification and control technology, defense nuclear waste and materials by-products management, and defense research and development. The waste also does not meet the definition of either spent nuclear fuel or high-level waste as provided in the *Waste Isolation Pilot Plant Land Withdrawal Act.*

FACILITY OPERATIONS

TRU waste is generated at the REDC in the hot cells of Buildings 7920 and 7930 (1) and in the support laboratories. The Building 7920 hot cells are used for four activities: target fabrication (cells 1-3), transuranium element processing (cells 4-7), analytical sample collection/storage (cell 8), and waste handling (cell 9). The layout of the Building 7920 hot cells is shown in Figure 2. Target fabrication consists of a series of mechanical operations to produce actinide targets for irradiation in the High Flux Isotope Reactor (HFIR) located at REDC. A number of quality assurance inspections are also performed as part of target fabrication.

Transuranium element processing is conducted on irradiated targets from the HFIR and on Mark 42 target assemblies irradiated at SRS (1). Processing operations includes dissolution of irradiated targets, separation of actinide elements from impurities and fission products, and separation of actinide elements from each other.

One hot cell is used to collect and store liquid samples for analysis in laboratories located elsewhere in Building 7920 (1). Samples are collected from various steps in the transuranium element processing operations to evaluate isotope content and purity. One hot cell is used for solid waste handling. Waste items from throughout the hot cell bank are transferred to this hot cell via an in-cell conveyor. Individual waste items are size-reduced if possible (e.g., plastic is melted, glass is crushed), segregated by activity (TRU vs. LLW, RH TRU vs. CH TRU) and by hazardous constituents (mixed vs. non-mixed). The waste management expert/operator is trained in RCRA requirements and therefore able to distinguish mixed from non-mixed waste. Waste items are then loaded into 1-gallon cans and these cans are then loaded into tie-bags (approximately 16 cans per bag). The waste items are then removed from the hot cells and loaded into containers.

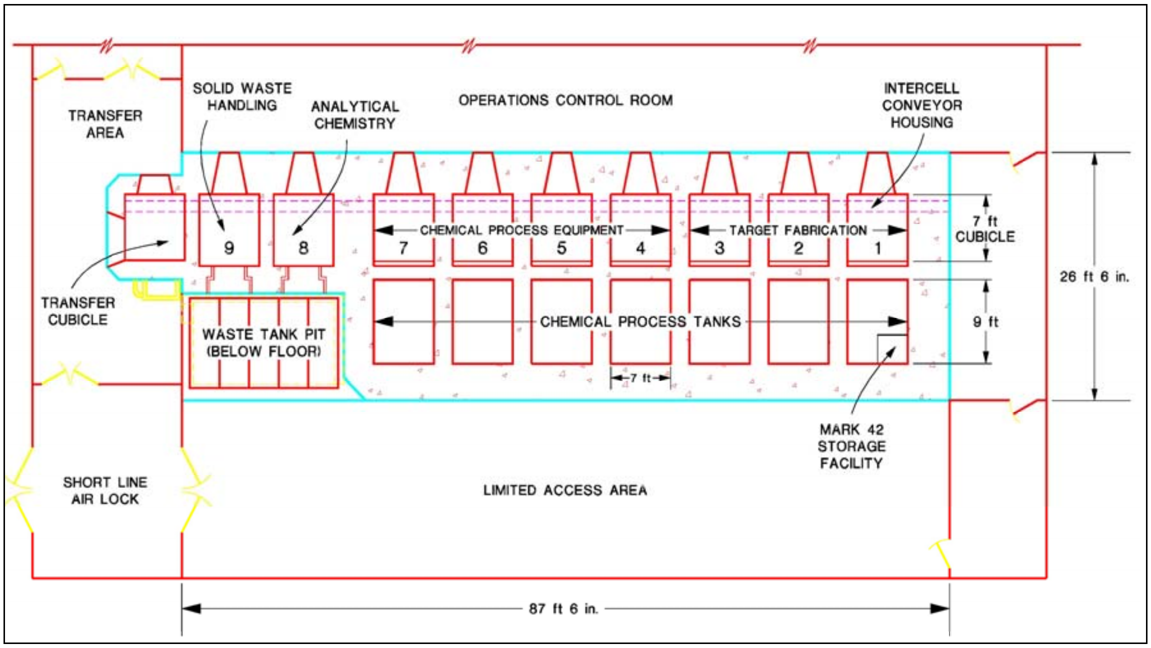

Fig. 2. Layout of Building 7920 Hot Cells

The Building 7930 hot cells are used for further processing of Californium from the transuranium element processing operations conducted in the Building 7920 hot cells, fabrication of Cf-252 neutron sources, packaging and shipping these sources, and recovery of Cm-248 from Cf-252 decay (3). The layout of the Building 7930 hot cells is shown in Figure 3. Californium processing includes pressurized ion exchange, pressurized extraction chromatography, resin loading-calcination, and oxalate precipitation-calcination. Cf-252 source fabrication includes pellet pressing, annealing, and rolling and swaging. Packaging of sources includes loading, assembly, and welding of capsules and shipping packages, X-ray examination, helium leak testing, decontamination of the capsules and packages, and assaying the contents of the packages.

Current and recently past TRU waste generation rates from Buildings 7920 and 7930 total approximately 14 m^3 per year for CH TRU waste and 7 m^3 per year for RH TRU waste. These estimates include TRU waste generated from the hot cells as well as the alpha and analytical laboratories. These estimates also include both mixed and non-mixed TRU waste. However, the vast majority of RH TRU waste generated at these buildings is non-mixed (1).

The WIPP WAP requires generator/storage sites to delineate waste streams for the purpose of TRU waste characterization. The WIPP WAP defines a waste stream as, "… waste material generated from a single process or from an activity that is similar in material, physical form, and hazardous constituents" (NMED 1999, pp. B-2). In accordance with WIPP guidance, TRU waste streams are defined by the activity that generated the waste, the physical form of the waste, whether or not the waste stream is hazardous (i.e., mixed waste), and whether the waste stream is CH TRU or RH TRU.

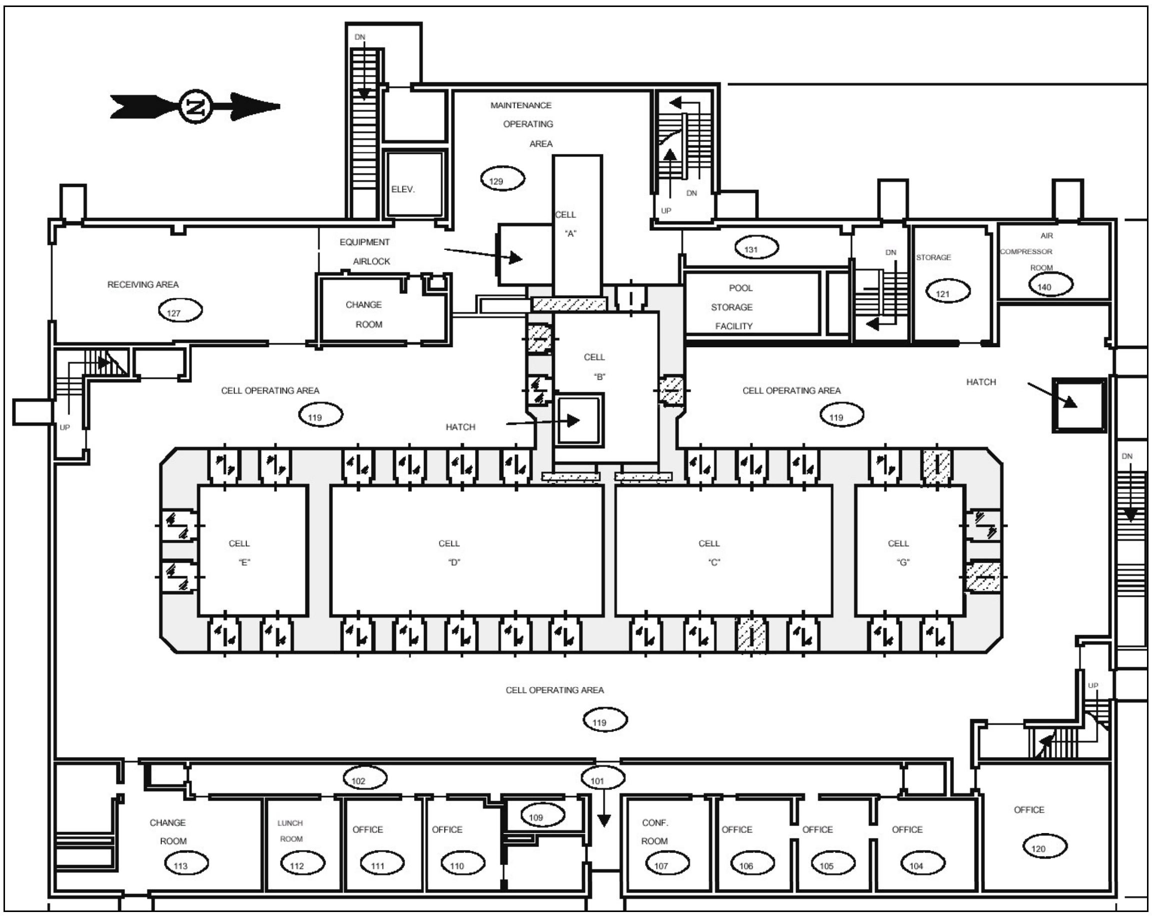

Figure 3. Layout of Building 7930 hot cells

TRU WASTE CHARACTERIZATION

TRU waste is generated during facility maintenance and operations in hot cells and support laboratories (1, 2) that may include routine or one-time operations to repair or replace equipment or to clean out facilities (4). In the hot cells of Building 7920, waste items are generated during target fabrication, isotope recovery/production, sample collection, and equipment maintenance activities. Individual waste items are transferred to cell 9 using an intercell conveyor and staged there until a waste packaging campaign is undertaken. Waste items are size reduced if possible (e.g., plastic is melted, glass is crushed) and segregated based on process knowledge and operator experience as CH TRU vs. RH TRU and mixed vs. non-mixed. Written procedures and check sheets will be implemented to document the process and ensure that no prohibited items are present in the waste (5). Activities may include certain activities to address prohibited items (e.g., solidify residual liquids, puncture sealed containers). Sharp objects or surfaces are taped or otherwise covered to ensure inner containers are not punctured. Waste items are then placed in 1-gallon cans and plastic tie-bags (approximately 16 cans per tie-bag). As waste is placed into cans, the information is documented on container log sheets.

RCRA Hazardous Waste Constituents

Based on a review of hot cell operating procedures for target fabrication, isotope recovery/production, waste management, and Material Safety Data Sheets for commercial products, the waste stream is non-mixed (5). Most of the reagents used in hot cell operations are various acids, bases, and salts. Waste management procedures restrict the presence of free liquids in TRU waste. In addition, all liquid wastes generated in the hot cells are collected in process waste tanks where actinide recovery are conducted before being disposed as liquid low-level waste (3, 4, 6). Therefore, the D002 and D003 EPA hazardous waste numbers do not apply.

Certain operating procedures list hazardous constituents as reagents that could render a waste hazardous (see Procedure HCOP-0402-R03, TF-CL-1002/R3, CF-OP-4.0/R2) (7, 8). These hazardous constituents are acetone, hydrazine hydrate, and methanol. However, these chemicals are either reacted during use (hydrazine hydrate) or dried prior to disposal (acetone, methanol) and so are not present in the waste such that the waste would be rendered as hazardous. Specifically, hydrazine hydrate reacts with palladium to form a palladium coating on Cf-252 pellets and wire. Therefore, the U133 EPA hazardous waste number does not apply. Acetone and methanol are used as solvents; however, items are allowed to dry after degreasing and cleaning or after being used in ion exchange operations and prior to disposal. Because of this, the F003 EPA hazardous waste number does not apply because these solvents are no longer ignitable when dry and there are no other RCRA listed spent solvents present in this waste stream.

Radiological Characterization

Data from twenty-eight surface smears were utilized to provide initial information for the radionuclides in this waste stream. The cotton tip swab smears taken using the hot cell surfaces and waste items staged for disposal in Cell 9 of Building 7920. The initial twenty-eight samples were used to explore the profile of alpha-emitting radionuclides in the REDC waste stream (9). The smears were analyzed using gross alpha, alpha spectrometry and gamma spectrometry. The waste from the REDC hot cells and support laboratories contains significant activity contributions from curium (Cm) and californium (Cf), primarily from Cm-244 and Cf-252. Neither Cm-244 nor Cf-252 is TRU by WIPP definition due to decay half lives less than twenty years. Therefore, they are categorized as non-TRU alpha emitting nuclides. The smear analysis was intended to profile the alpha-emitting nuclides in the REDC waste. Therefore, no additional radiochemical separations were performed to quantify pure or nearly pure beta emitters, such as Sr-90, known to be present in the waste. Additional smears were taken and analyzed. The large content (approximately 69% of the total activity) of Cm-244 present in both data sets was used to correlate or approximate the pure or near pure beta emitters that would have been present in the twenty-eight smear samples taken earlier.

The chemical processing conducted in the hot cells at REDC provides elemental separation while the isotopic ratio within the elements only change as a function of radioactive decay. Therefore, additional process knowledge supplementing the data from the twenty-eight smears was applied from isotopic mass ratios of curium and californium estimated in Campaign 68 Rework and Campaign 69 targets (10). The plutonium isotopic mass ratio of the waste was determined from analysis performed on the liquid waste generated from the process over a three-year period. The derived isotopic distribution (Table I) for the radiological characterization of the TRU waste from the REDC hot cell operations and the support laboratories are used as the basis for the radiological modeling.

The approaches described below are from the first phase of a two-phase process being used for the radiological characterization of newly generated (NG) TRU waste generated from the hot cell and support laboratories at REDC. The first phase defines the isotopic distribution as determined from direct samples taken from both waste material and equipment staged for disposal in the hot cell at REDC and the development of waste characterization models based on that distribution. The models reflect the upper limits of concentration for the nuclides present in the isotopic distribution. The second phase will collect additional samples and data to examine and assess measurement uncertainties, confidence intervals, and estimate relative error using field instruments to determine nuclide quantities inside NG TRU waste packages.

Dependant upon the contact dose rate emitted from the waste, it will be categorized as contact handled (CH) transuranic (TRU) waste, less than or equal to 2mSv/hr (200 mr/hr) at contact, or remote handled (RH) transuranic waste, greater that 2mSv/hr (200 mr/hr) at contact. The same isotopic distribution is used across both the CH and the RH waste, with the only difference being the total quantity of nuclides present. The 2mSv/hr (200 mr/hr) dose rate includes both the gamma and neutron dose contributions for the isotopes present in the TRU waste generated at REDC. The presents of spontaneous fission emitters, such as Cf-252 and Cm-248, in addition to the high non TRU alpha content, provide challenges to more conventional assay approaches used at other Department of Energy (DOE), which have primary constituents of plutonium.

The waste matrix may be contained in a 3.8 liter (1 gallon) can or in a 0.1 cubic meter (3.7 cubic feet) poly bag package and can be either CH or RH, dependent on the combined gamma and neutron dose rate. The ORNL Waste Characterization Plan (11) provides guidance for several techniques to calculate the nuclide concentrations in packages of radioactive waste, once the isotopic distribution is determined. The CH TRU waste at the poly bag level utilizes Method # 4 of this Plan. Two poly bags usually fill one 208-liter (55-gallon) drum. A modified 208-liter (55-gallon) drum technique is used to quantify the nuclides inside the drum of both CH and RH TRU waste. The gamma or photon emitting nuclides present in the isotopic distribution derived from hot cells and support laboratories in their respective fractions are use as the source material for the model.

Table I. Isotopic Distribution of Newly Generated (NG) TRU Waste Generated from the Hot Cell and Support Laboratories at REDC

	Nuclide	Ave. Act. (Bq/total)	Ave. Act. (Ci/total)	Total Act. (%)	Gamma Act. (%)
1	Co-60	2.01E+03	5.43E-08	0.02%	0.31%
2	Sr-90	1.87E+06	5.05E-05	15.49%	
3	Zr-95	2.72E+04	7.35E-07	0.23%	
4	Ru-103	5.03E+04	1.36E-06	0.42%	
5	Ru-106	2.27E+05	6.15E-06	1.88%	34.56%
6	Ag-110m	3.79E+03	1.02E-07	0.03%	
7	Sb-125	1.33E+04	3.60E-07	0.11%	2.02%
8	Cs-134	1.93E+04	5.22E-07	0.16%	2.94%
9	Cs-137	2.53E+05	6.83E-06	2.10%	38.43%
10	Ce-141	1.07E+05	2.89E-06	0.89%	
11	Ce-144	3.45E+04	9.32E-07	0.29%	
12	Eu-152	7.39E+03	2.00E-07	0.06%	1.12%
13	Eu-154	3.65E+04	9.87E-07	0.30%	5.55%
14	Eu-155	2.64E+04	7.13E-07	0.22%	4.01%
15	Np-239	9.66E+03	2.61E-07	0.08%	
16	Pu-238[a]	5.10E+04	1.38E-06	0.42%	
17	Pu-239[a]	2.49E+04	6.74E-07	0.21%	
18	Pu-240[a]	4.39E+04	1.19E-06	0.36%	
19	Pu-241	6.09E+05	1.65E-05	5.05%	
20	Pu-242[a]	6.36E+02	1.72E-08	0.01%	
21	Am-241[a]	6.79E+04	1.84E-06	0.56%	10.32%
22	Am-243[a]	4.85E+03	1.31E-07	0.04%	0.74%
23	Cm-242	3.72E+04	1.01E-06	0.31%	
24	Cm-244	8.36E+06	2.26E-04	69.25%	
25	Cm-246[a]	5.83E+04	1.58E-06	0.48%	
26	Cm-248[a]	1.56E+02	4.22E-09	0.00%	
27	Cf-249[a]	7.47E+01	2.02E-09	0.00%	
28	Cf-250	3.17E+03	8.56E-08	0.03%	
29	Cf-251[a]	1.38E+01	3.74E-10	0.00%	
30	Cf-252	1.23E+05	3.32E-06	1.02%	
		Total Act. Ci.	3.26E-04	100.00%	
		Total Gamma Act.	1.78E-05		100.00%

[a] TRU nuclide by definition

Radiological dose rate of containers of TRU waste from REDC are modeled using MicroShield®, version 5.01, to calculate photon dose rates emitted by the 208-liter (55-gallon) drum used as the container. The MicroShield model is constructed assuming the radionuclides have a uniform distribution throughout the matrix. The respective fraction of the gamma emitters present in the isotopic distribution is applied to the total source strength of one-curie. The MicroShield model calculates the total dose rate based on 37 GBq (1Ci) of total gamma activity. The calculated dose rate from the model is divided into the total activity to determine a mSv/hr (mr/hr) to GBq (Ci) coefficient to be used to calculate the gamma emitting nuclides present in the loaded package. Once the content of gamma emitters present in the waste package are determined, the non-gamma emitting nuclides are calculated based on the ratios of those gamma emitters present in the total isotopic distribution.

RH TRU waste is packaged in non-WIPP approved 2.4 meters (8 feet) high cylindrical high density concrete cask with 0.11 meter (4.5 inches) and 0.30 meter (12 inches) wall thickness. The high-density concrete, by design, shields much of the gamma emitters, but has very little effect on the spontaneous fission neutrons emitted by Cf-252. A neutron model was developed to calculate the quantity of Cf-252 present in the RH TRU waste cask based on Monte Carlo Neutron/Photon (MCNP) modeling of a Cf-252 source positioned in several location inside an empty cask (12). The results generated by the MCNP model were verified by making field measurement using a 102 microgram Cf-252 source. The area with the highest neutron emissions is located on the loaded cask and the neutron dose rate measurement is recorded. A second neutron rate is measured and recorded from the opposite side of the cask. The two neutron dose rates are averaged to move the neutron source to the theoretical center of the RH TRU cask. The 102 microgram Cf-252 source was divided by the average dose rate from the both sides of the RH TRU cask to determine a dose rate to microgram conversion coefficient for the Cf-252. The neutron dose rate from a loaded RH TRU waste cask is multiplied by the conversion coefficient to calculate the Cf-252 content inside the cask. The fraction of Cf-252 to the total activity of the distribution is used to calculate the remaining quantities of the nuclides in the isotopic distribution.

Currently all TRU waste on the ORR requires repackaging and certification before being shipped to the WIPP. A TRU waste processing and repackaging facility is under construction by Foster Wheeler Environmental Corporation (FWEC). The FWEC facility will provide treatment of TRU waste sludge, sorting, volume reduction, and WIPP certification for legacy CH and RH TRU waste in storage.

PREPARATION OF A WIPP COMPLIANT PROGRAM FOR THE REDC TRU WASTE

The REDC is the primary generator of TRU waste on the Oak Ridge Reservation. The REDC is operated for the DOE Office of Science by the UT-B. The TRU waste that results from activities in the REDC is managed by the BJC for the DOE Office of Environmental Management. UT-B and BJC have been working together to develop an integrated certification program for disposal of REDC's TRU waste at the WIPP. The general roles are that UT-B is the waste generator and BJC is the WIPP Certification Authority and overall manager of the TRU waste program. As such, BJC will provide the main interface with the WIPP concerning certification of the TRU waste and maintain the key TRU waste programs documents.

UT-B and BJC have been performing waste management functions using the established ORR Waste Certification Program (i.e., the "Program") (13). The Program establishes the requirements and responsibilities for certification of waste (including TRU waste) for disposal. The Program allows for a graded approach to the implementation to achieve the most cost-effective method for certification and is typically based on the waste acceptance requirement of the receiving facility. The Program uses ORR-specific waste profiles (referred to as "Master Profiles") and waste management procedures to precisely describe how the general requirements are to be met.

To ensure that ORR TRU waste generators meet the WIPP Waste Acceptance Criteria (WAC), BJC has developed a series of TRU waste Master Profiles based on the requirements found in the WIPP WAC. The profiles reflect the requirements related to material description, chemical constituent limitations, radiological constituent limitations, physical parameter limitations, characterization parameters and methodology, prohibited items, packaging requirements, additional requirements, and required documentation. BJC issues "variances" to certain requirements in the Master Profiles that cannot be met until after the ORR has completed the development of the TRU waste program and the WIPP has concurred with the program requirements.

To support compliance with the ORR TRU Waste Certification Program and document compliance with the BJC Master Profile System, ORNL has established the ORNL Waste Certification Program (14). The ORNL Waste Certification Program consists of a series of program plans, guidance documents, and procedures that establish the responsibilities and requirements for TRU waste characterization, acceptance, and certification. To implement the requirements of the ORNL Waste Certification Program for TRU waste characterization, REDC has established guidance documents, training requirements, and waste characterization and packaging procedures for the TRU waste being generated in the hot cells of Buildings 7920 and 7930.

The BJC contracted the Benchmark Environmental Corporation (Benchmark) to help with the development of the WIPP program documents. Benchmark, BJC, and UT-B have developed a draft certification plan, quality assurance plan, and acceptable knowledge report as a baseline for REDC waste. Benchmark will continue to assist BJC and UT-B in developing program documentation in 2002 and 2003 to prepare for an anticipated audit by the WIPP in 2004 for newly generated CH-TRU waste.

CONCLUSION

The BJC and UT-B are working to develop a WIPP compliant certification program for the REDC TRU waste. The REDC waste provides unique challenges for conventional NDA applications due to the presents of Cm-244, approximately 69% of the total activity and Cf-252, approximately 1% of the total activity. The AK for the NG TRU waste utilizes the available information from the active production operations at REDC to determine the physical, chemical, and radiological characteristic of the waste generated from REDC. The certification program will use the combine efforts of UT-B and BJC to meet the WIPP criteria for disposal.

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