Alternative Radiological Characterization of Sealed Source TRU Waste for WIPP Disposal (LAUR-05-8776)

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ABSTRACT

The Offsite Source Recovery (OSR) Project at Los Alamos National Laboratory is now shipping transuranic (TRU) waste containers to the Waste Isolation Pilot Plant (WIPP) in New Mexico for disposal. Sealed source waste disposal has become possible in part because OSR personnel were able to obtain Environmental Protection Agency (EPA) and DOE-CBFO approval for an alternative radiological characterization procedure relying on acceptable knowledge (AK) and modeling, rather than on non-destructive assay (NDA) of each container. This is the first successful qualification of an "alternate methodology" under the radiological characterization requirements of the WIPP Waste Acceptance Criteria (WAC) by any TRU waste generator site. This paper describes the approach OSR uses to radiologically characterize its sealed source waste and the process by which it obtained certification of this approach.

INTRODUCTION

Beginning in the late 1950's, the federal government distributed radioactive materials for use in industrial, medical, and health effects applications and research. The Atomic Energy Act was amended in 1954 to promote the peaceful use of radioactive materials by civilians and allow the U.S. to assist foreign countries developing peaceful nuclear programs. These efforts resulted in relative widespread distribution of "batches" of materials containing Pu-238, Pu-239 and Am-241 to selected licensed manufacturers for production and distribution of sealed sources [Ref. 1]. Under the "Low-Level Waste Policy Amendments Act of 1985" [Ref. 2], Congress assigned to DOE the responsibility for the management and disposal of "Greater-than-Class C" waste, including actinide sealed sources. In response, DOE established the OSR Project with a mission to remove unwanted, excess sealed sources that were distributed into the public domain and move them to disposal or secure storage [Ref. 1, 3]. The events of September 11, 2001, heightened the urgency of this mission; in September 2002, Congress tasked the OSR Project with the recovery a large number of excess and unwanted actinide sealed sources by April 2004 [Ref. 4].

The OSR project uses three different versions of a WIPP-approved pipe overpack container (POC) for packaging, storing, and disposing of sealed sources (Fig. 1.). These containers, which serve multiple functions, allow the project staff to handle sources only one time during the recovery, storage, and disposal operations. The POC, a very robust container, consists of a standard stainless steel 55-gallon drum containing a 1/4-inch thick stainless steel pipe centered within cane fiberboard dunnage or polyethylene neutron shielding, depending on the type of configuration. Within the stainless steel pipe, additional high density polyethylene may be used for neutron attenuation. Inside of this, each sealed source is placed in a DOT-compliant stainless steel special form capsule (Fig. 2), which has ½-inch thick stainless steel walls. Finally, most sources as manufactured are doubly encapsulated in tantalum and stainless steel. As a result of this required packaging and the resultant shielding of the radiological materials contained, assay of sealed source containers is problematic at best and likely to result in significant underestimation of the isotopic quantities involved, if any radioactive material can be detected at all.

Fig. 1. Example Pipe Overpack, S100 configuration

Fig. 2. Special form capsules

TECHNICAL APPROACH

Development of Radiological Characterization Approach

EPA has specified that radiological characterization information not generated under a WIPP-approved QA program be qualified by one four methods listed in 40 CFR 194.22(b) [Ref. 6], including qualification under an NQA-1-equivalent program, confirmatory testing, and peer review conducted compatible with NUREG 1297, *Peer Review for High-Level Nuclear Waste Repositories* [Ref. 7]. Project personnel determined that, with existing AK information of high quality for each of three types of actinide sealed sources, peer review was the desired methodology for qualifying such information.

First, OSR developed a sealed source radiological characterization process for the three different major TRU radionuclide sources: Pu-239; Pu-238; and Am-241. For each source type, Project personnel reviewed shipping and ownership transfer data, national database information, NRC and state regulatory licenses, TRU batch material records from source manufacturers, manufacturer catalogues, material type designations, fabrication documents and drawings, and other records to develop isotopic distributions and quantify associated errors for each of the ten WIPP-tracked radionuclides, as well as a few others that were potentially significant on a mass basis. The resulting radionuclide mass ratios and associated errors are developed and quantified in three calculation reports published by the OSR Project [Ref. 8, 9, 10] and are shown in Tables I-III below.

For Pu-239 sealed sources, the primary information used in the development of the ratios are [Ref 8] the NMMSS database (a listing last updated in 1985 of almost all of the known Pu sealed sources manufactured in the US) [Ref. 11], material type-based radiological distributions from internal LANL reports, and impurity concentrations of Am-241, fission products, and U reported for Pu-238 material. The vast majority of Pu-239 sealed sources were material type 52, which is essentially weapons-grade Pu. The resulting (pre-decay) isotopic ratios are shown in Table I. The following factors were included in uncertainty calculations: uncertainties from Pu measurement by gamma spectroscopy, uncertainty in total Pu measurements from calorimetry during manufacture, variability in the amount of Pu-240 across different material types, variability in the different material types used in source manufacture, and errors in impurity measurements. Uncertainties range from 2.39% for Pu-239, the primary isotope, to 245% for Pu-242.

Table I. Summary of Representative Radionuclide Distribution and Associated Uncertainties for Pu-239 Sealed Sources

Radionuclide	Ci of Radionuclide per Gram (g) of Pu in Source	g of Radionuclide per g of Pu in Source	Total Uncertainty
Pu-238	2.56E-03	1.48E-04	149.8%
Pu-239	5.86E-02	9.32E-01	2.4%
Pu-240	1.50E-02	6.50E-02	23.7%
Pu-241	2.53E-01	2.44E-03	63.9%
Pu-242	1.44E-06	3.62E-04	118.8%
Am-241	9.84E-04	2.50E-04	96.2%
U-234	2.15E-11	3.00E-09	51.4%
U-235	9.32E-13	3.75E-07	51.4%
U-238	5.77E-11	1.50E-04	51.4%
Cs-137	2.04E-05	2.35E-07	81.5%
Sr-90	1.83E-05	1.03E-07	81.5%
Source: Ref. 8, 12			

For Pu-238 sealed sources, the primary information used in determining the radiological distribution included numerous batch analytical data sheets from Pu-238 obtained from the original manufacturer and NMMSS data on individual sources [Ref. 9]. Both an 80 wt% Pu-238 and a 90 wt% Pu-238 composition were observed, but the NMMSS review showed that almost 98% of the sealed sources were of the 80 wt% composition [Ref. 11]. The batch analytical data sheets provided the composition of Pu-238 to -242 isotopes with a relative standard deviation of only 0.26 – 0.38% for the Pu-238 isotope [Ref. 13]. These data sheets also provided impurity masses for Am-241 and U, as well as a gamma activity per unit mass of the Pu. The U impurity was assumed to have the isotopic distribution of depleted U. The resulting distribution is shown in Table II. Uncertainties were calculated considering the following: a manufacturing uncertainty typical for gravimetric measurements; Pu measurement uncertainty based on calorimetric measurements; isotopic distribution uncertainties from measurement by gamma spectroscopy; variability in NMMSS data; and errors in the impurity measurements for U [Ref. 9]. As shown in Table II, the resulting uncertainties ranged from 1.46% for Pu-238, the primary isotope, to 96.15% for Am-241.

Table II. Summary of Representative Radionuclide Distribution and Associated Uncertainties for Pu-238 Sealed Sources

Radionuclide	Mass Fraction	Grams of Isotope per gram of Pu-238	Ci of Isotope per Gram of Pu Metal	Total Uncertainty
Pu-238	8.03E-1	1.00E+00	1.39E+01	1.46%
Pu-239	1.61E-1	2.01E-01	1.01E-02	3.22%
Pu-240	2.63E-2	3.28E-02	6.05E-03	3.70%
Pu-241	6.90E-3	8.59E-03	7.17E-01	23.0%
Pu-242	2.33E-3	2.90E-03	9.25E-06	54.3%
Am-241	2.84E-4	3.53E-04	9.84E-04	96.15%
U-234	3.40E-9	4.24E-09	2.15E-11	51.4%
U-235	4.25E-7	5.30E-07	9.32E-13	51.4%
U-238	1.70E-4	2.11E-04	5.77E-11	51.4%
Cs-137	2.31E-7	2.88E-07	2.04E-05	81.5%
Sr-90	1.33E-7	1.66E-07	1.83E-05	81.5%
Source: Ref. 9, 12				

Analytical data sheets generated during manufacturing of Am-241 batches were also used as a primary source of information for the calculation of mass ratios for this source type. In this case, the data sheets accounted for almost 75% of the entire mass of material used in the manufacture of all Am-241 sealed sources [Ref. 14]. The resulting distribution is shown in Table III. Sources of uncertainty included uncertainty in the amount of Am placed in each source, Am calorimetry measurement errors, impurity measurement errors (including gamma activity), and variability in the analytical sheet data [Ref. 10]. The resulting uncertainties ranged from 1.4% for Am-241, the primary isotope, to 98% for Cs-137 and Sr-90 impurities.

Table III. Summary of Representative Radionuclide Distribution and Associated Uncertainties for Am-41 Sealed Sources

Radionuclide	Grams (g) of Isotope per g of Am-241	Ci of Isotope per g of Am-241	Total Uncertainty
Am-241	1.00E+00	3.47E+00	1.4%
Pu-238	2.06E-06	3.56E-05	55.6%
Pu-239	2.76E-03	1.79E-04	54.4%
Pu-240	5.57E-04	1.23E-04	54.5%
Pu-241	3.98E-05	4.14E-03	54.4%
Pu-242	1.20E-05	4.75E-08	55.6%
U-234	1.15E-11	7.26E-14	74.8%
U-235	1.44E-09	3.14E-15	74.8%
U-238	5.73E-07	1.95E-13	74.8%
Cs-137	7.80E-10	6.88E-08	98.0%
Sr-90	4.48E-10	4.83E-08	98.0%
Sources: Ref. 10, 12			

Software Development

The next step in developing OSR’s radiological characterization process involved developing a method for accounting for decay and production processes in the sources, many of which are more than 40 years old. These processes have a minimal effect on the quantity of Pu-239 in each source type, but are more important for Pu-238 and Am-241 sources [Ref. 15]. OSR opted to use an existing decay calculation software package, RadDecay, with a custom-built program that would interface between the radiological data present for each source in an MS Access database and the RadDecay software. This custom-built program, NucDecay v1.0 [Ref. 16], simply converts Access input files into files compatible with RadDecay and does the reverse for output files generated by RadDecay. To produce the input files, the Access database applies the radionuclide distribution listed above applicable to the specific source type and data supplied by the user (primary isotope quantity and manufacture date) and generates the input file, which comprises values for all 10 WIPP-reportable radionuclides. A data input screen and output file are shown in Fig. 3 and Table IV, respectively.

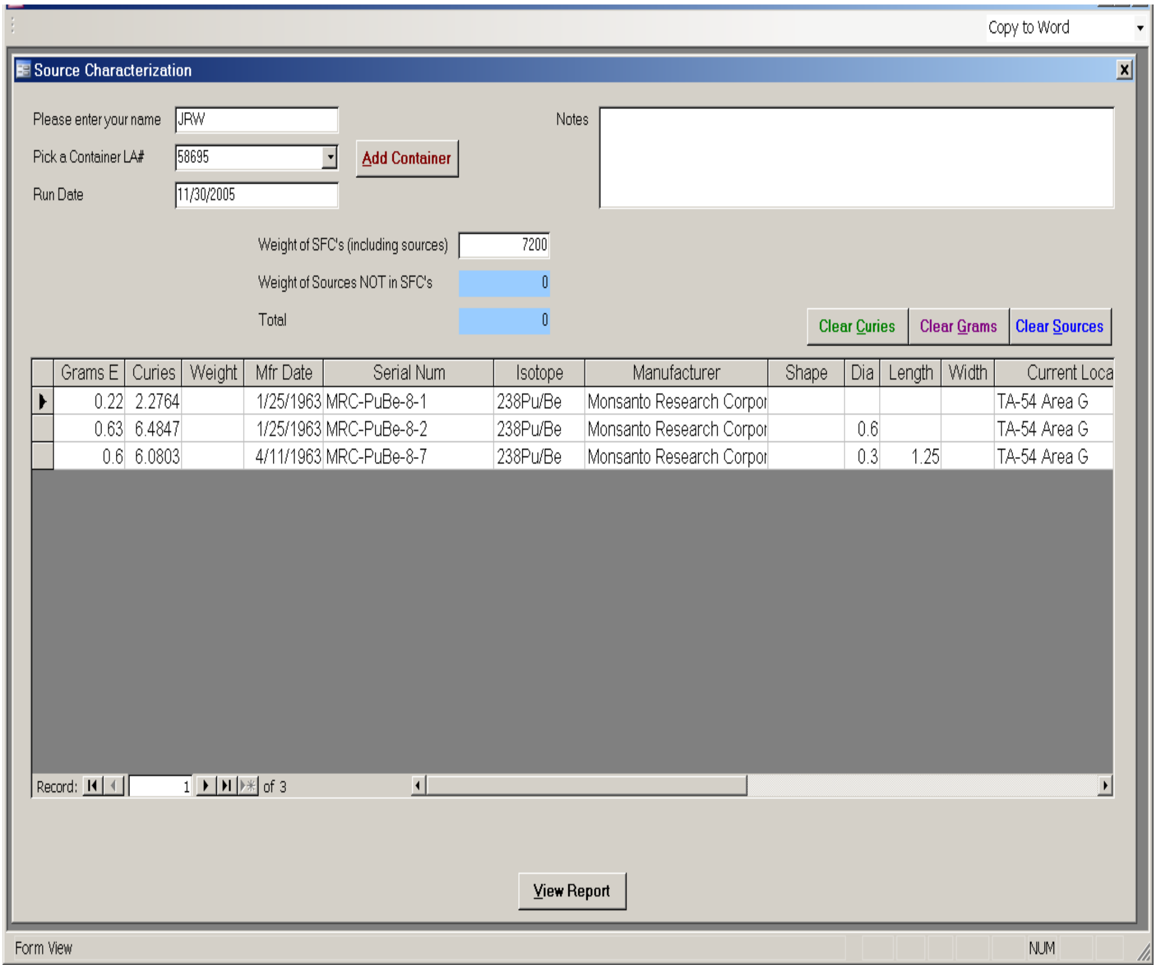

Fig. 3. Example access input screen

Table IV. Example Container Characterization Report (Output File)

Total Container Content With Uncertainty Applied -- All Sources								
Nuclide	Adjusted Content (Grams)	Adjusted Activity (Ci)	Adjusted Activity (TBq)	FGE (2s)*	PECi	W	% Type A Limit	TRU Alpha Activity (Ci)
137Cs	6.69E-06	5.89E-04	2.18E-05	0.00E+00	0.00E+00	6.51E-07	1.09E-05	0.00E+00
233U	4.30E-08	4.20E-10	1.55E-11	5.12E-08	1.31E-10	1.22E-11	3.82E-13	0.00E+00
234U	4.25E-03	2.68E-05	9.93E-07	0.00E+00	0.00E+00	7.73E-07	2.44E-08	0.00E+00
235U	5.51E-02	1.21E-07	4.47E-09	3.61E-02	0.00E+00	3.33E-09	0.00E+00	0.00E+00
238Pu	1.08E-02	1.86E-01	6.88E-03	1.86E-03	1.69E-01	6.16E-03	6.89E-04	1.86E-01
238U	1.13E-02	3.83E-09	1.42E-10	0.00E+00	0.00E+00	9.71E-11	0.00E+00	0.00E+00
239Pu	4.54E+01	2.86E+00	1.06E-01	4.62E+01	2.86E+00	8.86E-02	1.06E-02	2.86E+00
240Pu	3.67E+00	8.43E-01	3.12E-02	9.48E-02	8.43E-01	2.62E-02	3.12E-03	8.43E-01
241Am	1.62E-01	5.64E-01	2.09E-02	4.02E-03	5.64E-01	1.88E-02	2.09E-03	5.64E-01
241Pu	2.20E-02	2.29E+00	8.46E-02	6.55E-02	4.48E-02	7.28E-05	2.08E-03	0.00E+00
242Pu	4.91E-02	1.95E-04	7.21E-06	6.06E-04	1.77E-04	5.75E-06	7.22E-07	1.95E-04
90Sr	3.63E-06	5.00E-04	1.85E-05	0.00E+00	0.00E+00	5.80E-07	6.18E-05	0.00E+00
Other	NA	NA	NA	NA	NA	NA	NA	NA
Totals	4.89E+01	6.32E+00	2.34E-01	4.64E+01	4.26E+00	1.33E-01	1.74E-02	4.24E+00

Total alpha n**Ci/g** **5.89E+05**

*** All uncertainties are 1 s, except FGE, which is 2 s**

Performed By: **Operator 1** **Date:** **3/31/2005**

Reviewed By: ______________________________ **Date:** ____________

Approved: ____________________

OSR developed this software under the LANL TRU Waste Characterization Program (TWCP) QA program. However, it never began to be used until the TWCP was replaced at LANL by DOE CBFO’s Central Characterization Program (CCP). As a result, the software had to be requalified under CCP’s QA program, including the procedure “CCP TRU Software Quality Assurance,” CCP-QP-022 [Ref. 17]. Although OSR had developed all of the documentation required under this program for vendor software (such as a verification and validation document), completion of a Software Evaluation Checklist demonstrated that the software qualified as Commercial Off-the-Shelf software, minimizing the number of additional requirements.

Peer Review

Concurrent with the development of radionuclide distributions for each source type and software to account for decay and production process, an independent Peer Review Panel was convened in accordance with the guidance in NUREG 1297 [Ref. 7] to qualify existing AK radiological data as described in 40 CFR 194.22(b) [Ref. 6]. The panel evaluated the AK information provided by OSR to determine whether the information proposed for use in the characterization process described by LANL was acceptable for the radiological characterization of sealed source waste in compliance with the WIPP WAC [Ref. 1, 18]. Review of the software was not part of the panel’s scope. During the review, OSR provided examples of “primary” documents such as the NMMSS database, source certificates, source shipping data sheets, and fabrication documents that alone contain sufficient information to determine the primary isotope, quantity, and manufacture date for each source. OSR also provided examples of “secondary” documents that have at least one of these pieces of information, including source and device markings, the national NRC Device Registry, manufacturer catalogues, drawings, and NRC licenses.

In the final Peer Review Panel Report [Ref. 18], the panel concluded that the specific documentation and information identified constituted “adequate documentation for determining the radionuclide type, the radionuclide content/activity and either the date of manufacture or other more conservative date (for purposes of decay correction).” The report also stated that “AK results far surpass any that could currently be generated using NDA,” in terms of accuracy and uncertainty. Finally, the panel recommended that the proposed characterization approach not be used for sources that were deliberately physically altered, had missing or illegible documentation, were irradiated (such as sources present in reactors), or had severely inconsistent documentation.

Certifying Program Modifications

OSR project personnel drafted a new procedure, CCP Offsite Source Recovery Project Sealed Source Radiological Characterization, CCP-TP-101 [Ref. 19], describing the radiological characterization process. This procedure contains the definition of sufficient AK as developed during the peer review process [Ref. 18]. Documents that do not fall within the “sufficient AK” definition in this procedure are not allowed to be used as the sole AK sources defining inputs for radiological characterization. The procedure also requires identifying the major isotope and its quantity in each source, determining a manufacture date, and using this data as input to the software, which applies the appropriate distribution and accounts for decay and other as described previously.

In addition to adding the new procedure, some procedures owned by the certifying program, the CCP, were changed to accommodate the proposed characterization methodology. These included the following:

- CCP Quality Assurance Project Plan (CCP-PO-001) [Ref. 20]
- CCP Transuranic Waste Certification Plan (CCP-PO-002) [Ref. 21]
- CCP Acceptable Knowledge Documentation procedure (CCP-TP-005) [Ref. 22]
- CCP Project Level Data Validation and Verification procedure (CCP-TP-001) [Ref. 23]
- CCP Reconciliation of Data Quality Objective Reconciliation and Reporting Characterization procedure (CCP-TP-002) [Ref. 24]
- CCP Transuranic Waste Certification and WWIS Data Entry (CCP-TP-030) [Ref. 25]
- CCP-LANL Interface Document (CCP-PO-012) [Ref. 26].

No changes were required to CCP's TRAMPAC document, CCP-PO-003, as this document already covered the use of POCs.

Regulatory Approval

Once the peer review panel approved the types of documentation OSR had collected for each source as adequate for identifying and quantifying the major isotope for each source, OSR sought DOE approval for the overall radiological characterization process. DOE-Carlsbad Office agreed to allow the OSR process to be examined by EPA during the LANL site's normal audit process. The approvals of both entities were formally obtained in writing in June, 2005 [Ref. 27].

Programmatic Implications

The AK-based approach used by OSR is consistent with a current Class 3 permit modification sought by DOE-CBFO for the WIPP site. In an NOD response dated September 22, 2005 [Ref. 28], the permittees describe the proposal to "use acceptable knowledge for waste analysis if it is sufficient" and to conduct sampling and analysis if the AK is not sufficient. This idea of sufficiency determination is similar in spirit to the definition of "sufficient AK" present in the OSR radiological characterization procedure [Ref. 19].

The Remote-Handled TRU Waste Characterization Program Implementation Plan [Ref. 29], however, more directly discusses the use of AK information for radiological characterization purposes. It contains an Acceptable Knowledge Procedure for Remote-Handled TRU Waste (Attachment A) that allows for qualification of AK information by peer review, among the other methods allowed in 40 CFR 194.22 [b] [Ref. 6], and also contains requirements that must be met in the case where there is insufficient AK information to address the radiological characterization Data Quality Objectives, which are defined elsewhere in the main document. In the procedure, the primary characterization method is AK that is qualified under 40 CFR 194.22 [b], consistent with the OSR radiological characterization approach outlined in this paper. The other allowable qualification methods are confirmatory testing, demonstration that AK information was collected under an NQA-1 equivalent program, and corrobating data.

CONCLUSION

The OSR project successfully developed and obtained approval for radiological characterization of its transuranic waste stream by AK plus calculations, rather than assay of every container. The AK information involved was qualified in accordance with 40 CFR Section 194.22 [b] by peer review. This approach may benefit waste generators of other difficult-to-assay waste streams, including RH-TRU waste streams.

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