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Radiological Characterization Technical Report for Curium-244 Sealed Source Transuranic Debris Waste for the Off-Site Source Recovery Project at Los Alamos National Laboratory

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ACRONYMS

AK	Acceptable Knowledge
DOE	Department of Energy
FGE	Fissile Gram Equivalent
HFIR	High Flux Isotope Reactor
IDL	Instrument Detection Limit
MCNP	Monte Carlo N-Particle (code)
MDA	Minimum Detectable Activity
NMMSS	Nuclear Materials Management Safeguards System
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
OSRP	Off-Site Source Recovery Project
REDC	Radiochemical Engineering Development Center
SF	Spontaneous Fission
SFC	Special Form Capsule
SRS	Savannah River Site
TRU	Transuranic
US	United States
WAC	Waste Acceptance Criteria
WIPP	Waste Isolation Pilot Plant

ABSTRACT

This document describes the development and approach for the radiological characterization of Cm-244 sealed sources for shipment to the Waste Isolation Pilot Plant. The document uses the extensive documentation and information on the nuclear material content of each source (mass or activity of material used) and information and data on the radionuclide distributions within the nuclear material. This information allows complete characterization without the need for additional measurements.

The radionuclide uncertainties were developed from acceptable knowledge (AK) information on the radiological contents of the source materials used in the manufacture of the sources. The AK information used in the characterization of Cm-244 sealed sources has been qualified by the peer review process, which has been reviewed and accepted by the Environmental Protection Agency.

1 INTRODUCTION

Sealed sources were manufactured for a variety of uses, including nuclear batteries, smoke detectors, neutron flux generators, and threshold detectors, and for a number of applications. Over the years a significant number of these sealed sources that were manufactured for a wide range of applications have ceased to be used and have been declared to be excess. Under Public Law 99-240 titled the “Low-Level Waste Policy Amendments Act of 1985” (Reference 1), Congress assigned the United States (US) Department of Energy (DOE) the responsibility for the management and disposal of “Greater-than-Class C” waste as defined in the Nuclear Regulatory Commission’s (NRC’s) 10 CFR Part 61 regulations (Reference 2), which includes actinide sealed sources. In response to Public Law 99-240, DOE established the Off-Site Source Recovery Project (OSRP) for the specific purposes of recovering, managing, and disposing of excess or unwanted actinide sealed sources. This radiological characterization report addresses Cm-244 sealed sources. The Cm-244 used in the manufacture of the sealed sources was produced by both Savannah River Site (SRS) and Oak Ridge National Laboratory (ORNL) reactors and purified in chemical processing facilities at ORNL. This material was offered for sale to commercial entities or other DOE laboratories where the sealed sources were fabricated. The Cm-244 sealed sources were intended to be used primarily as heat generation sources and alpha particle sources. The sealed sources addressed in this report were declared to be excess and were no longer useful. Further details on this waste stream will be provided in CCP-AK-LANL-008, *Central Characterization Project Acceptable Knowledge Summary Report for Los Alamos National Laboratory Off-Site Source Recovery Project Sealed Sources, Waste Stream: LA-OS-00-01.001, LA-OS-00-03, LA-OS-00-04* (Reference 3).

Because no facility other than the Waste Isolation Pilot Plant (WIPP) currently offers safe and secure permanent disposal capabilities, it is intended that the actinide-bearing sealed sources be disposed of as transuranic (TRU) waste at WIPP. As such, the sealed source waste packages

must meet the WIPP Waste Acceptance Criteria (WAC) (Reference 4). These criteria include the requirement for the radiological characterization of waste packages to identify the quantities and types of radionuclides in the package before they are disposed of. The effort described in this report provides the necessary radiological characterization of actinide sealed source waste packages to be in compliance with the WIPP WAC.

2 BACKGROUND

2.1 Cm-244 Production

In 1959, SRS became involved in the production of transplutonium elements created by the irradiation of Pu-239. The Cm-244 is produced by successive neutron captures, as shown in Figure 2-1.

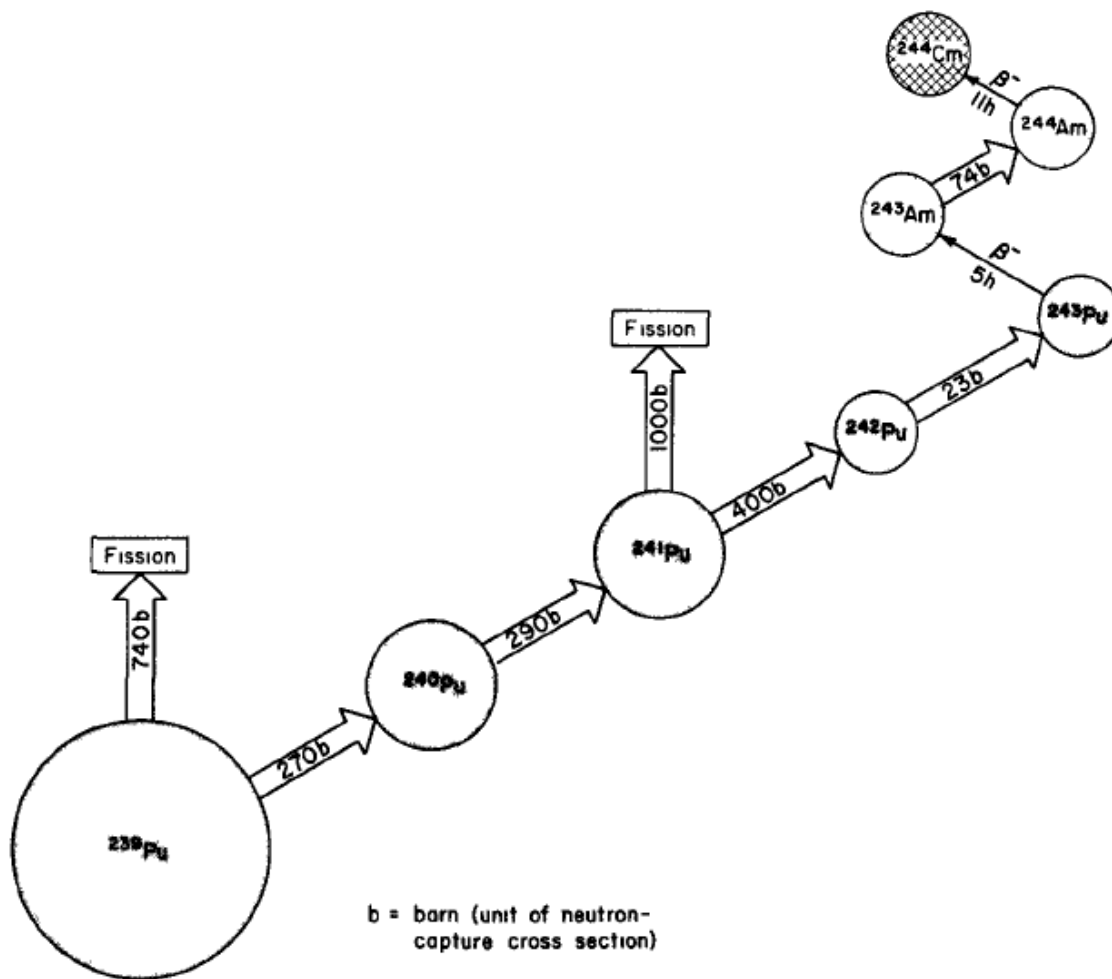


Figure 2-1. Curium-244 production scheme by neutron capture.

The program was expanded in the early 1960s to produce larger quantities of Cm-244; a two-stage irradiation approach was followed by subsequent irradiations of plutonium with heavier isotopes and americium isotopes. This production scheme is shown in Figure 2-2.

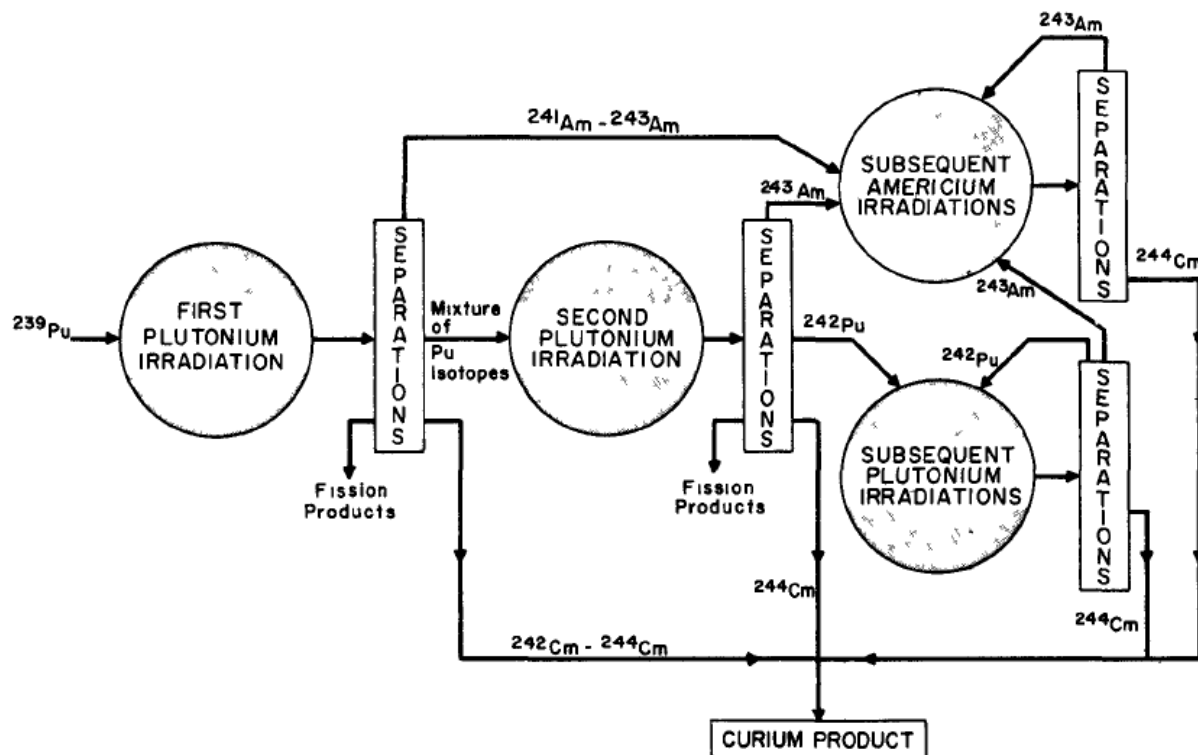


Figure 2-2. Production diagram for Cm-244.

The irradiations were completed at both SRS and ORNL in the High Flux Isotope Reactor (HFIR), with the final separations and purifications taking place at ORNL.

2.2 Cm-244 Source Descriptions

The sizes and configurations of the Cm-244 sources varied with the application intended for the source. Alpha sources were offered by commercial suppliers in sizes ranging from 1 μCi up to 1 mCi. This was especially true for the Cm-244 source supplied as heat sources for a variety of applications. Depending on the heating requirement for a given application, the Cm-244 content could be as large as a curie or more.

3 TECHNICAL APPROACH

The approach proposed for the radiological characterization of Cm-244 sealed source waste is to use the extensive documentation and information on the nuclear material content of each source (mass or activity of material used) and information and data on the radionuclide distributions within the nuclear material. This information allows for complete characterization without the need for additional measurements.

The WIPP WAC (Reference 4) requires that, as a minimum, the masses and activities of the following 10 radionuclides (if present) be reported:

Am-241	U-233
Pu-238	U-234
Pu-239	U-238
Pu-240	Sr-90
Pu-242	Cs-137

In addition to the above 10 radionuclides, other radionuclides are required to be reported that, in the aggregate, constitute not less than 95% of the total radiological hazard based on the A_2 values given in US Department of Transportation (DOT) packaging regulations, 49 CFR, *Transportation*, Part 173, *Shippers—General Requirements for Shipments and Packagings*, Section 435, Table of A1 and A2 Values for Radionuclides (Reference 5). Also, the *Contact-Handled Transuranic Waste Authorized Methods for Payload Control (CH-TRAMPAC)* (Reference 6) requires that 95% of the decay heat be accounted for in the report of the radionuclides in a package, plus any radionuclide that contributes >1% of the total.

The relative concentrations of the radionuclides in the source material were evaluated to determine the reporting requirement for parameters, such as fissile gram equivalent (FGE), decay heat, and radiological hazard (Reference 7). The evaluation showed that Cm-244 is required to be reported because of its contribution to the radiological hazard, the FGE, and decay heat. In addition, Cm-245 is also required to be reported because of its contribution to FGE. Also reported are the other TRU radionuclides, Cm-246, Cm-247, Cm-248, and Am-243, which are found as impurities in the curium sources.

The characterization approach involves four steps.

1. Determine the primary radionuclide and the quantity of the primary radionuclide in the source based on acceptable knowledge (AK) information and data specific to the individual sources.
2. Apply the specific radionuclide impurity distribution that has been developed from AK information for the identified primary radionuclide material in the source.

3. Perform the decay correction of the source material from the time of manufacture to the time of shipment for disposal.
4. Summarize the radionuclide content of all sources loaded into the special form capsule (SFC) and ultimately into a 55-gallon drum.

The uncertainties developed from AK information regarding the source materials are applied to the summed activities in the drum.

Three specific pieces of AK information are required as input for the radiological characterization of sealed source waste:

- the type of source (identification of the primary radionuclide),
- the radionuclide content of the source (curies or grams), and
- the date of manufacture.

The impurity radionuclide distributions and associated uncertainties were developed from AK information on the radiological contents of the source materials used in the manufacture of the sources. The Cm-244 source material was produced and purified at ORNL before being distributed to source manufacturers. The stockpile of Cm-244 source material was produced and periodically repurified to remove the ingrowth of impurity radionuclides. The source material was sampled and analyzed for radionuclide content to meet source material impurity requirements. These batch analyses were used to develop average impurity levels in the materials. The variability in the measured impurity levels was used to develop the uncertainties in the source content and radionuclide distribution. The AK information used in the characterization of Cm-244 sealed sources has been qualified by the peer review process (Reference 8), which has been reviewed and accepted by the Environmental Protection Agency (EPA).

4 DEVELOPMENT OF REPRESENTATIVE RADIONUCLIDE DISTRIBUTIONS

This section presents the derivation and quantification of the WIPP-required radionuclide distributions that will be applied to the primary radionuclide. The Cm-244 source material is typically 86–92 wt% Cm-244, with the remainder of the material containing other curium isotopes and trace impurities of other radionuclides resulting from the separation and purification processes. The trace impurities are primarily plutonium isotopes. To estimate the radionuclide distribution in the Cm-244 product material, analytical records were retrieved from the Radiochemical Engineering Development Center (REDC) at ORNL, where the Cm-244 source material was purified before being shipped.

The original stockpile of Cm-244 source product material was analyzed on February 20, 1979. As the material was supplied to meet future demands, the original source material was re-purified to remove the ingrowth radionuclides. A total of eight ORNL analytical results were retrieved, representing the future purifications that followed the original production run. Figure 4-1 provides an example of one of the analytical results sheets, showing the original curium composition of the source material and subsequent composition in 1994 based on decay calculations.

Other analytical results sheets show mass spectrometry analysis results subsequent to the original mass spectrometry measurements on the original source material, along with radiochemical analyses of the impurities in the material following the purification step. Figures 4-2 and 4-3 provide an example of the second results datasheet.

The reported values in the analytical results for the impurities in the Cm-244 source materials included

- Am-241,
- Am-243,
- Pu-238,
- Pu-240, and
- Cs-137.

The number of reported values for the above impurities is shown in Table 4-1.

**ANALYTICAL DATA FOR ^{244}Cm PRODUCT
(BATCH: CMP-576, FRACTION: 9H79Cm-PROD)**

Table 1. Mass Spectrometer Analysis
Date of Analysis: February 20, 1979

<u>Isotope</u>	<u>Atom %</u>	<u>Weight %</u>
^{244}Cm	92.94	92.89
^{245}Cm	1.09	1.09
^{246}Cm	5.80	5.84
^{247}Cm	0.11	0.11
^{248}Cm	0.06	0.06

Table 2. Mass Spectrometer Analysis
Decayed to: April 11, 1994

<u>Isotope</u>	<u>Atom %</u>	<u>Weight %</u>
^{244}Cm	88.108	88.028
^{245}Cm	1.837	1.843
^{246}Cm	9.767	9.838
^{247}Cm	0.186	0.188
^{248}Cm	0.101	0.103

Table 3. Gross α and Pulse-height Analysis
Date of Analysis: August 13, 1993

<u>Alpha Energy (Nuclide)</u>	<u>Abundance</u>
5.15 MeV (^{240}Pu)	0.30 %
5.80 MeV (^{244}Cm)	99.70 %

Volume: 10.0 mL
Gross α : $2.93\text{E}+10$ Bq/mL
Gross N_{f} : $6.93\text{E}+5$ cts/m/mL

Figure 4-1. ORNL analytical data for Cm-244 product (Batch: CMP-576, Fraction 9H79 Cm-Prod.).

**ANALYTICAL DATA FOR FOR ^{248}Cm PRODUCT
(BATCH: 4P61Cm-PROD)**

Table 1. Mass Spectrometer Analysis
Date of Analysis: January 8, 1986

<u>Isotope</u>	<u>Atom %</u>	<u>Weight %</u>
^{244}Cm	91.141	91.079
^{245}Cm	1.360	1.365
^{246}Cm	7.310	7.365
^{247}Cm	0.121	0.122
^{248}Cm	0.068	0.067

Average Molecular Weight = 244.232^a

Weight % ^{244}Cm = 90.957^a

Weight % ^{244}Cm = 88.821^a

^aAdjusted to March 31, 1986.

Table 2. Mass Spectrometer Analysis
Decayed to: August 30, 1996

<u>Isotope</u>	<u>Atom %</u>	<u>Weight %</u>
^{244}Cm	87.268	87.182
^{245}Cm	1.956	1.962
^{246}Cm	10.504	10.581
^{247}Cm	0.174	0.176
^{248}Cm	0.098	0.099

Table 3. Radiochemical Analyses
Date of Last Actinide Separation: August 15, 1996
Date of Analysis: September 6, 1996

<u>Alpha Energy (Nuclide)</u>	<u>Abundance</u>
5.80 MeV (^{244}Cm)	100.0 %
Volume: 10.4 mL	
Gross α : 5.90E+10 Bq/mL	205.22 mg of ^{244}Cm
Pu(TTA) α : 4.82E+06 Bq/mL (100.0 %, 5.15 MeV)	5.97 mg of ^{240}Pu
Gross Np: 1.50E+06 c/m/mL	241.57 mg of ^{244}Cm
γ -Scan: 7.90E+05 Bq/mL	64.78 μg of ^{241}Am
2.60E+06 Bq/mL	3.67 mg of ^{243}Am
8.00E+06 Bq/mL	0.010 μg of ^{239}Np
5.50E+05 Bq/mL	0.05 μg of ^{144}Ce

Figure 4-2. ORNL analytical results for Cm-244 product (Batch: 4P61 Cm-Prod.).

Table 4. Radiochemical Impurities¹
Date of Analysis: September 6, 1996

Nuclide	Bq/mL ¹	μg/mg ²⁴⁴ Cm
⁹⁵ Zr	1.10E+04	< 7.02E-07
⁹⁵ Nb	5.50E+03	< 1.92E-07
¹⁰³ Ru	6.60E+03	< 2.80E-07
¹⁰⁶ Ru	6.40E+04	< 2.65E-05
¹²⁵ Sb	1.80E+04	< 2.35E-05
¹³⁴ Cs	6.70E+03	< 7.09E-06
¹³⁷ Cs	8.00E+03	< 1.26E-04
¹⁴⁰ La	9.80E+03	< 2.41E-08
¹⁴⁰ Ba	2.30E+04	< 4.31E-07
¹⁴¹ Ce	1.70E+04	< 8.17E-07
¹⁵² Eu	2.70E+04	< 2.10E-04
¹⁵⁴ Eu	2.20E+04	< 1.14E-04
¹⁵⁵ Eu	5.10E+04	< 1.50E-04

¹Minimum Detectable Activities

Table 5. Chemical Impurities^a
Date of Analysis: January 7, 1986

Element	μg/g Cm ₂ O ₃	Element	μg/g Cm ₂ O ₃
Al	5	P	200
B	5	Pb	200
Ba	1	S	50
Ca	30	Si	200
Cl	30	Sn	500
Co	10	Ta	≤5
Cr	3	Ti	10
Cu	1	V	1
Fe	100	Zn	140
K	3	Y	1
Mg	3	La	1
Mn	1	Ce	3
Mo	5	Pr	1
Na	100	Nd	30
Ni	20	Heavy R.E. ^b	≤7800

^aMeasured by spark source mass spectroscopy.

^bHeavy rare earth elements.

Figure 4-3. Second page of ORNL analytical results for Cm-244 product (Batch: 4P61 Cm-Prod.).

Table 4-1. Number of Reported Values from the Eight Analytical Sheets

Radionuclide	Number of Reported Values
Am-241	6
Am-243	7
Pu-238	3
Pu-239	0
Pu-240	8
Pu-242	0
Cs-137	2

The average concentrations of the actinides relative to Cm-244 were determined from these analytical results. Because Pu-239 and Pu-242 were not reported, their potential contribution to the impurities was evaluated by assuming that the plutonium isotopic distribution would be similar to that measured in the curium targets in the Radiochemical Engineering Development Center (REDC) at ORNL (Reference 9).

The Pu-238 scaling factors from the HFIR campaigns in the REDC are given in Table 4-2. The two plutonium isotopes of Pu-239 and Pu-242 were scaled to the reported values for Pu-238 from the analytical sheets described above.

Table 4-2. Average Pu-238 Scaling Factors and Relative Pu-239 and Pu-242 Concentrations from Curium Target Materials in the REDC

HFIR/HFIR Scaling Factors:				
Radionuclide	Ci/Ci	g/g Pu-238	SA	Initial g/g Cm-244
Pu-238	2.87E+00	-	1.73E+01	4.35E-05
Pu-239	5.34E-04	5.12E-02	6.29E-02	2.23E-06
Pu-242	1.04E-03	1.58E+00	3.97E-03	6.87E-05

For comparison, the relative concentrations of Pu-239 and Pu-242 from the decay of Am-243 and Cm-246 were calculated, as given in Table 4-3.

Table 4-3. Pu-239 and Pu-242 from the Decay of Am-243 and Cm-246

Parent	lambda (yr ⁻¹)	Daughter	20-Year Decay (g/g Cm-244)
Am-243	9.39E-05	Pu-239	3.69E-05
Cm-246	1.46E-04	Pu-242	3.64E-04

The resulting concentrations from the decay calculations were significantly larger than the concentrations that were estimated from the Pu-238 concentrations in the reported impurities. Accordingly, the estimated initial impurity concentrations for Pu-239 and Pu-242 were neglected and only the Pu-239 and Pu-242 produced by the decay of Am-243 and Cm-246 were considered.

Only two values were reported for Cs-137 on the analytical sheets. In addition, five minimum detectable activity (MDA) values were reported. As an upper estimate of the Cs-137 impurity level, the MDA values were assumed to be real values and were averaged with the two reported values. Although Sr-90 was not reported, it was assumed that the Sr-90 impurity level would equal the Cs-137 concentrations.

Uranium isotopes were not measured and not reported because any uranium impurities would have been removed in the multiple processing of the curium source product material. Accordingly, the uranium isotope impurities were considered to be negligible.

The contributions of Cs-137 (and Sr-90) to the impurity levels from the spontaneous fission (SF) of Cm-244, Cm-246, Cm-248, and Pu-239 were also considered. The ultimate concentration of the fission products depends on the fission time due to buildup; consequently, different fission times were considered. Table 4-4 shows the results of the calculations for the buildup of Cs-137 as a function of the fission time.

Table 4-4. Cs-137 Contribution from Spontaneous Fission

	Cs-137 from SF of Cm-244	Cs-137 from SF of Cm-246	Cs-137 from SF of Cm-248	Cs-137 from SF of Pu-240	Total Cs- 137 from SF	Initial Cs-137	Total Cs-137	% from SF
Years	Ci/Ci Cm-244	Ci/Ci Cm-244	Ci/Ci Cm-244	Ci/Ci Cm-244	Ci/Ci Cm- 244	Ci/Ci Cm-244	Ci/Ci Cm-244	
1	2.42E-09	1.92E-10	7.22E-12	9.50E-15	2.62E-09	1.93E-07	1.96E-07	1.3%
2	4.69E-09	3.79E-10	1.43E-11	2.75E-14	5.09E-09	1.89E-07	1.94E-07	2.6%
3	6.83E-09	5.61E-10	2.12E-11	5.34E-14	7.41E-09	1.84E-07	1.92E-07	3.9%
20	2.71E-08	3.10E-09	1.17E-10	1.31E-12	3.04E-08	1.25E-07	1.55E-07	19.6%
38	3.00E-08	4.89E-09	1.85E-10	3.30E-12	3.51E-08	8.25E-08	1.18E-07	29.9%
39	2.99E-08	4.97E-09	1.88E-10	3.41E-12	3.51E-08	8.06E-08	1.16E-07	30.3%
40	2.98E-08	5.05E-09	1.91E-10	3.53E-12	3.50E-08	7.88E-08	1.14E-07	30.8%

At a period of 20 years, the contribution of Cs-137 from SF is ~20% of the impurity level; however, because the Cs-137 impurity levels are so small, it was considered reasonable to neglect this contribution.

Another source of fission products is the fast neutron fission induced by neutrons emitted during SF. This source was assessed (Reference 10) by the use of the Monte Carlo N-Particle code (MCNP) 5. From these analyses it was concluded that fission products from neutron-induced

fission are negligible compared with the fission products from SF, which as shown above are negligible compared with the impurity levels already found in the Cm-244 product material.

Table 4-5 summarizes the curium isotope distribution for the source material analyzed on February 20, 1979 (Reference 11).

Table 4-5. Summary of the Curium Isotopic Distribution in the Source Material Analyzed on February 20, 1979

	Atom Fraction	Lambda (yr ⁻¹)	Relative Activity	Activity Relative to Cm-244 Activity, Ci/Ci of Cm-244
Cm-244	0.9294	3.83E-02	3.56E-02	1.00E+00
Cm-245	0.0109	8.15E-05	8.89E-07	2.50E-05
Cm-246	0.058	1.46E-04	8.46E-06	2.38E-04
Cm-247	0.0011	4.44E-08	4.89E-11	1.37E-09
Cm-248	0.0006	2.04E-06	1.23E-09	3.45E-08

Table 4-6 summarizes the radionuclide distribution of “fresh” curium source material on the date of source manufacture.

Table 4-6. Radionuclide Distribution of Fresh Curium Source Material

Radionuclide	Concentration	Units
U-233	<LLD	Ci/Ci of Cm-244
U-234	<LLD	Ci/Ci of Cm-244
U-235	<LLD	Ci/Ci of Cm-244
U-238	<LLD	Ci/Ci of Cm-244
Pu-238	9.28E-06	Ci/Ci of Cm-244
Pu-239	0.00E+00	Ci/Ci of Cm-244
Pu-240	5.86E-05	Ci/Ci of Cm-244
Pu-242	0.00E+00	Ci/Ci of Cm-244
Am-241	9.96E-06	Ci/Ci of Cm-244
Am-243	4.77E-05	Ci/Ci of Cm-244
Cm-244	1.00E+00	Ci/Ci of Cm-244
Cm-245	*	Ci/Ci of Cm-244
Cm-246	*	Ci/Ci of Cm-244
Cm-247	*	Ci/Ci of Cm-244
Cm-248	*	Ci/Ci of Cm-244
Cs-137	1.97E-07	Ci/Ci of Cm-244
Sr-90	1.97E-07	Ci/Ci of Cm-244

* The concentration of the curium isotopes will be determined by decay calculations for the fresh source on the date of source manufacture.

5 DETERMINATION OF THE QUANTITY OF NUCLEAR MATERIALS IN SEALED SOURCES

In general, significant documentation and information exist on sealed sources manufactured in the US, including the original production, transportation, and source control documents. However, for individual sealed sources, variability exists in the quantity and extent of the documentation available for use in the characterization of the sealed source. Because sealed sources are manufactured to meet user specifications, strict adherence to procedures under the oversight of quality assurance programs of the day ensured that these sources and their associated production documents were prepared with a high degree of care and accuracy. The nature of the source production and the historically successful functioning of these sources to meet their intended purposes support this observation. The nuclear material contents of the sources were carefully controlled to meet the specifications imposed by the end user on the power output, neutron flux, impurity levels, etc.

After a source is declared excess, but before it is recovered, all available information and documentation on the source is retrieved, reviewed, and evaluated to determine if the information is adequate relative to the identity of the primary radionuclide in the source and the quantity of the radioactive material in the source (including both primary and secondary information sources). This information may be derived from a set of documents that are considered *primary* information sources in that they contain all of the information necessary to establish the source type (primary radionuclide) and the quantity of radionuclide material in the source. The primary information source documents include the

- source certificates,
- source shipping datasheet,
- Nuclear Materials Management Safeguards System (NMMSS) database, and
- source fabrication documents.

Secondary information source documents that also address one or more of these three pieces of information are the

- source and device markings (usually etched or stamped on the source),
- NRC registry,
- source manufacturer's sales catalogues,
- source drawings,
- source physical dimensions (measured at the time of recovery), and
- NRC licenses.

Typically, the secondary documents may require the use of two or more separate source documents to identify both the primary radionuclide and the radionuclide content in the source.

Generally, only one piece of documentation is required from the list of primary AK information to identify the radionuclide and quantity in a sealed source. Primary AK information is derived from the

- NMMSS,
- shipping records,
- source certificate, and
- fabrication documents.

This primary AK information is unique to a specific sealed source as identified by a unique serial number. The NMMSS database is available in the public domain and includes information on the

- source manufacturer,
- serial number,
- material type (primary radionuclide),
- weight of the element,
- weight of the isotope,
- date of manufacture,
- ownership,
- possession, and
- NRC license number.


The remaining three primary AK information sources typically show the primary radionuclide, the quantity of material, and the serial and model number of a specific sealed source. Examples of a licensing document and a source certificate are shown in Figures 5-1 and 5-2.

If these information sources are not available, then reliance is placed on the secondary AK information. One document from the secondary AK information may identify the primary radionuclide in the sealed source, and a second may identify the quantity of material in the source. The secondary AK information includes the

- source markings,
- NRC registry,
- manufacturer's sales catalogues,
- sealed source drawings,
- unique physical descriptions, and
- miscellaneous other documents.

TRC Form 12-1
08/04

Page 1 of 2


Department of State Health Services

GENERAL LICENSE ACKNOWLEDGEMENT

Pursuant to the Texas Radiation Control Act and Title 25 of the Texas Administrative Code (25 TAC), Chapter 289, an acknowledgement is hereby issued to the general licensee listed below for the possession, use and transfer of radioactive material in device(s) obtained under the provisions of 25 TAC Section 289.251(g). The general licensee is subject to all applicable rules, regulations and orders of the Department of State Health Services (Agency) now or hereafter in effect and to any requirements specified in the conditions listed below.

<p style="text-align: center;">GENERAL LICENSEE</p> <p>1. Name EMAG LLC 2. Contact Person RICHARD MCCOY 3. Address 1715 FOURTH STREET GRAHAM TX 76450</p>	<p>This acknowledgement is issued in response to a 2-year fee payment</p> <p>Received: March 5, 2007</p> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%;">4. Acknowledgement Number G01588</td> <td style="width: 50%;">Amendment Number 06</td> </tr> </table> <p style="text-align: center;">PREVIOUS AMENDMENTS ARE VOID</p> <p>5. License Expiration Date March 31, 2009</p>	4. Acknowledgement Number G01588	Amendment Number 06
4. Acknowledgement Number G01588	Amendment Number 06		

6. Device Manufacturer	7. Device Model No.	8. Device Serial No.	9. Source Serial No.	10. Radionuclide	11. Activity*
A. NDC Systems	105	6002	0599LM	Cm-244	200 mCi
B. NDC Systems	105	6015	0420LM	Cm-244	200 mCi
C. NDC Systems	105	6018	0600LM	Cm-244	200 mCi
D. NDC Systems	105	6019	0601LM	Cm-244	200 mCi
E. NDC Systems	105	6035	0597LM	Cm-244	200 mCi
F. NDC Systems	105	6162	3526LM	Cm-244	100 mCi

* Ci - Curies; mCi - millicuries; μ Ci - microcuries

12. A. Radioactive material possessed under the authorization of the general license granted at Section (§) 289.251(g) of Title 25 of the Texas Administrative Code (25 TAC) shall be possessed and used at and records pertaining to the possession of that radioactive material shall be maintained for inspection by the Agency at:

Site Number	Location
000	Graham - 1715 Fourth Street

B. Portable and/or mobile devices may also be used at temporary sites, not under exclusive Federal jurisdiction, throughout Texas.

13. The general licensee shall comply with the applicable provisions of 25 TAC §289.201, §289.202(w) and (xx), §289.204, §289.205, §289.251, and §289.257.

14. The radioactive material possessed under the authorization of the general license granted in 25 TAC §289.251(g) shall be used only for the purpose for which the device containing the radioactive material was approved as specified in the Registry of Radioactive Sealed Sources and Devices.

15. Within 60 days of receipt, the general licensee shall submit a completed GLA Self-Evaluation Form (provided by the Agency annually).

Figure 5-1. Example of source licensing document.

1/17/92

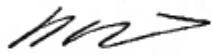
Radioactive source test report

SHEET 1 OF 1

product code CL008411		description CURTUM-244 L.E.G DISC SOURCE C/E E/15/3266		customer order no 392198 1		BSI/ISO classification C64344	
AI item no Z89691 1		nominal activity 250mCi 9.25GBq		customer AMERSHAM CORPORATION C/O CIZZON CORP CUSTOMS BROKER 9650 W FOSTER AVENUE CHICAGO ILLINOIS 60656 AEC L USA		special form certificate no GB/144/S	
source model no				recommended working life 10 YEARS			

serial no batch no	measurement 1		measurement 2		test D	test L	test A
	*EMISSION	date		date			
1874LM	16.4 E+06	02/09/90			02/01/92	02/01/92	02/01/92
2146LM	18.8 E+06	11/11/91			24/09/91	07/11/91	07/11/91
2235LM	18.7 E+06	30/12/91			19/12/91	23/12/91	23/12/91
→ 2236LM	17.3 E+06	30/12/91			19/12/91	23/12/91	23/12/91
2237LM	16.7 E+06	30/12/91			19/12/91	23/12/91	31/12/91
2238LM	18.3 E+06	30/12/91			19/12/91	23/12/91	23/12/91
2239LM	18.9 E+06	30/12/91			19/12/91	23/12/91	23/12/91
2240LM	16.2 E+06	30/12/91			19/12/91	23/12/91	31/12/91
2241LM	17.2 E+06	30/12/91			19/12/91	23/12/91	23/12/91
2243LM	18.3 E+06	30/12/91			19/12/91	23/12/91	23/12/91

notes
*EMISSION PHOTONS PER SECOND PER STERADIAN 18KEV
DATE OF DESPATCH 13 JANUARY 1992

signature  position **Prod
Sip.** date **03/01/92**

*this classification complies with BS5288:1976, which is in agreement with ISO2919
(see overleaf for definition and description of tests)

Amersham International plc
Amersham Laboratories
White Lion Road
Buckinghamshire England HP7 9LL

Telephone (0494) 544000
Fax (0494) 543243
Cables Activity Amersham
Telex 83141 ACTIVA G



Figure 5-2. Example of sealed source certificate.

Many sources will have etched markings on the source itself. Often these markings will indicate the primary radionuclide and the quantity of the material in the source. The source markings may identify the model number of the sealed source to allow it to be traced to the NRC registry or to fabrication information for that model of sealed sources. The NRC Registry of Radioactive Sealed Sources is available in the public domain and typically identifies the

- source manufacturer,
- manufacturer model numbers,
- primary radionuclide, and
- maximum quantity of radioactive material in the different source models.

Manufacturers' sales catalogues will generally include information on the sealed source models that have been manufactured, such as the

- model number,
- primary radionuclide,
- quantity of radioactive material in the source, and
- physical dimensions of the sealed source.

In many cases, the availability of the physical dimensions on a particular source model will allow measurements to be made in the field at the time of recovery to confirm the source manufacturer, model, and source content.

If further information is needed for the source, photographs may be taken if the source has a unique physical shape that allows the source model number to be identified. Once the model number is identified, other documents can then be used to identify the primary radionuclide and the content of the sealed source. Figures 5-3 through 5-5 provide examples of the NCR registry, a source photograph, and a manufacturer's sales catalogue.

REGISTRY OF RADIOACTIVE SEALED SOURCES AND DEVICES
SAFETY EVALUATION OF SEALED SOURCE
(AMENDED IN ITS ENTIRETY)

NO.: CA0471D101B

DATE: February 6, 2006 **PAGE:** 1 of 7

SEALED SOURCE TYPE: Thickness Gauge

MODEL: 105, 107

MANUFACTURER/DISTRIBUTOR:

NDC Infrared Engineering, Inc.
5314 North Irwindale Avenue
Irwindale, CA 91706

SEALED SOURCE MODEL DESIGNATION:

Amersham Model CLCL,
Amersham AMC.D3 (was Model AMC.66)
Amersham Model AMCL

ISOTOPE:

Curium 244 (**Model CLCL**)
Americium 241 (**Model AMC.D3**)
Americium 241 (**Model AMCL**)

MAXIMUM ACTIVITY:

250 millicuries
150 millicuries
100 millicuries

LEAK TEST FREQUENCY:

Six (6) months for Cm-244 sources
Three (3) years for Am-241 (**AMC.D3**)
Six (6) months for Am-241 (AMCL)

PRINCIPAL USE: X-ray Fluorescence (U)

CUSTOM SOURCE: _____ YES X NO

Figure 5-3. Example of an NRC Registry of Sealed Sources.



Figure 5-4. Example of sealed source photograph.



Curium-244

γ and Primary X-ray Sources

Curium-244 incorporated in a ceramic enamel, sealed in a welded monel capsule with brazed beryllium window; the active component is recessed into a tungsten backing.

Nominal activity*		A	B	Typical photon output in 17keV Pu LX-rays	Product code
GBq	mCi	mm	mm		
0.37	10	10.8	7	0.8×10^6	CLC10990
1.11	30	10.8	7	2.4×10^6	CLC11564
3.7	100	10.8	7	7.8×10^6	CLC11562
7.4	200	10.8	7	15.0×10^6	CLC11377

* Tolerance $\pm 10\%$

Nominal activity*		A	B	Typical photon output in 17keV Pu LX-rays	Product code
GBq	mCi	mm	mm		
0.37	10	8	4	0.8×10^6	CLC11932
1.11	30	8	4	2.4×10^6	CLC11284
3.7	100	8	4	7.8×10^6	CLC11933

* Tolerance $\pm 10\%$

Recommended working life: 10 years

Quality control

Wipe Test I

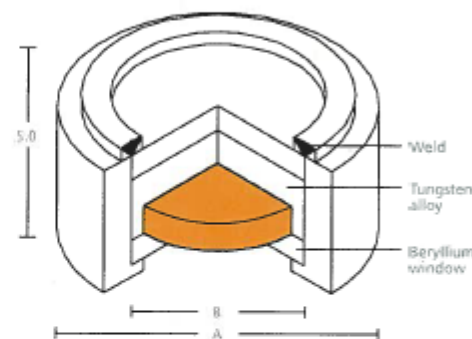
Immersion Test II

Bubble Test III

Neutron emission

All Curium-244 sources emit $\sim 3.6 \times 10^5$ n/s per GBq due to spontaneous fission and (α ,n) reactions with the low atomic number elements (e.g. Si, Al, O) in the active material.

VZ-3069



Capsule dimensions and safety performance testing

Overall diam. 'A' mm	Active diam. 'B' mm	Window thickness 'C' mm	Safety performance testing	
			ANSI/ISO classification	IAEA special form
8.0	4	1	C64343	YES
10.8	7	1	C64344	YES

Figure 5-5. Example of a manufacturer's sales catalogue.

6 CHARACTERIZATION OF CM-244 SOURCES IN WASTE DRUM

After the Cm-244 sealed sources are recovered, they are loaded into a 55-gallon drum or they are placed into a special form capsule (SFC), which, in turn, is loaded into a 55-gallon drum. For the characterization of the drum, an Excel spreadsheet is used for the information input and the subsequent calculations for the characterization of the drum, including the quantities of all radionuclides estimated to be in the drum; derived parameters, such as TRU concentration and decay heat; and the uncertainties in these values. All of the documentation on each source is reviewed, and the required information for the characterization of the source is input to a Data Input Sheet in the Excel workbook (Reference 12). The required information is the source identity, the activity quantity of the primary radionuclide (Cm-244), the date of manufacture for the source, and the estimated total weight of the SFC and sources. Figure 6-1 shows an example of the data input sheet for the Cm-244 characterization spreadsheet.

Container Characterization Report Cm-244 Sources

Data Input Sheet

NDA/Radiological Characterization BDR#
Run Date
Waste Stream Designation
Container Number
Estimate Waste Weight, kg

LA11-OSR-CH-00X	
11/15/2011	
Cm-244 Sealed Sources	
1234	
7.7	

Source ID	Manufacture Date	Source Contents, Curies	Source Contents, Grams
91-191	1/1/1985		0.4
91-192	1/1/1985	3.00E-02	
91-193	1/1/1985	3.00E-02	
91-194	1/1/1985	3.00E-02	
91-195	1/1/1985	3.00E-02	
91-196	1/1/1985	3.00E-02	
91-197	1/1/1985	3.00E-02	
91-198	1/1/1985	3.00E-02	
91-199	1/1/1985	3.00E-02	
91-200	1/1/1985	3.00E-02	

Figure 6-1. Example of a Characterization Input Sheet.

Each source is decay corrected from the assumed manufacture date to the run date entered into the input sheet. Specifically, the radionuclides of Cm-244, Cs-137, Sr-90, and Pu-238 are decay

corrected. Curium-246 is decay corrected to yield the contribution of Pu-242 to the source contents. Similarly, the Am-243 is decay corrected to yield the contribution of Pu-239.

The spreadsheet calculates the quantities, both activity and mass, for all of the radionuclides in the drum and associated uncertainties in these values. The derived parameters of TRU, FGE, PE-Ci, decay heat, and total activity are also calculated.

Figure 6-2 provides an example of the characterization summary report for a drum of Cm-244 sources.

Container Characterization Report

Cm-244 Sources

Version 1.0

NDA/Radiological Characterization BDR#

Run Date

Waste Stream Designation

Container Number

Net Waste Weight

LA11-OSR-CH-00X	
11/15/2011	
Cm-244 Sealed Sources	
1234	
7.7	kg

Nuclide	% Type A Limit	Activity (Ci)	Grams	FGE	PECi	Watts	Uncertainty in Curies	Uncertainty in Grams
U-233	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
U-234	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
U-238	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Cm-244	2.18E+00	1.18E+01	1.44E-01	1.30E-02	6.21E+00	4.12E-01	8.34E-02	1.02E-03
Cm-245	4.29E-04	1.03E-03	5.92E-03	8.88E-02	1.10E-03	3.41E-05	1.03E-04	5.93E-04
Cm-246	4.08E-03	9.78E-03	3.14E-02	0.00E+00	1.04E-02	3.21E-04	1.20E-04	3.85E-04
Cm-247	6.98E-08	5.66E-08	6.03E-04	3.02E-04	5.44E-08	1.80E-09	5.67E-09	6.05E-05
Cm-248	2.64E-04	1.42E-06	3.31E-04	0.00E+00	5.28E-06	1.83E-07	1.43E-07	3.32E-05
Pu-238	9.17E-05	2.48E-04	1.43E-05	1.62E-06	2.25E-04	8.20E-06	1.09E-04	6.32E-06
Pu-239	4.49E-07	1.21E-06	1.93E-05	1.93E-05	1.21E-06	3.76E-08	1.60E-07	2.55E-06
Pu-240	2.24E-02	6.04E-02	2.62E-01	5.90E-03	6.04E-02	1.88E-03	1.36E-03	5.92E-03
Pu-242	1.80E-07	4.86E-07	1.22E-04	9.18E-07	4.42E-07	1.43E-08	4.89E-08	1.23E-05
Am-241	1.22E-04	3.29E-04	9.47E-05	1.77E-06	3.29E-04	1.10E-05	1.20E-04	3.45E-05
Am-243	1.12E-03	1.57E-03	7.79E-03	1.00E-04	1.57E-03	5.06E-05	1.36E-04	6.74E-04
Sr-90	4.18E-05	3.39E-06	2.46E-08	0.00E+00	0.00E+00	3.93E-09	1.05E-06	7.63E-09
Cs-137	6.49E-06	3.51E-06	3.98E-08	0.00E+00	0.00E+00	3.88E-09	1.09E-06	1.24E-08
Totals	2.21E+00	1.19E+01	4.53E-01	1.08E-01	6.28E+00	4.15E-01		

TRU Alpha Activity Concentration

TRU Alpha Activity

Total Pu-239 Equiv Activity

Total Pu-239 Fissile Gram Equiv

Total Decay Heat

Value	(one Sigma)	Units
9.52E+03	1.84E+02	nCi/g
7.33E-02	1.42E-03	Ci
6.28E+00	4.46E-02	Ci
1.08E-01	8.91E-03	g
4.15E-01	2.95E-03	W

(Print Name)

Signature

Date

RC Operator Review

2nd RC Operator Review

Figure 6-2. Example of a Characterization Summary Report.

7 UNCERTAINTY ANALYSIS

This section discusses the uncertainty in the reported values of the radionuclides in the drums containing the Cm-244 sources. The radionuclide inventory of each sealed source is calculated based on the manufacturer's reported Cm-244 activity and the radionuclide distributions derived from sample data, as described in Section 4.0. Sources of uncertainty are summarized in Table 7-1.

Table 7-1. Summary of Sources of Uncertainty

Source of Uncertainty	Treatment
Manufacturer's uncertainty in mass or activity of source	$\pm 5\%$ per source, with 50 sources per drum
Uncertainty in curium isotopics	Based on typical uncertainties for mass spectrometry measurements
Uncertainty in other radionuclide impurities	Standard deviation of the batch sample data
Instrument uncertainty	Included as one source of variability in the sample data.
Uncertainty in decay time from last purification to shipping date	$\pm 10\%$

7.1 Manufacturer's Source Material Uncertainty

The Cm-244 sources were manufactured by a number of different manufacturers. Reference 13 is a product catalog for one manufacturer. This catalog specifies that, for various alpha sources (including Cm-244), the source material will be determined to within 2%–5%. A value of 5% is assumed for each source. Each drum is assumed to contain at least 50 sources of approximately equal size. Therefore, the drum uncertainty due to the manufacturer's uncertainty is $\sim 5/\sqrt{50} \% = 0.71\%$. It is assumed that sources from other manufacturers have a similar level of uncertainty.

7.2 Radionuclide Impurity Distribution Uncertainty

It is assumed that the curium sources come from the same feedstock at ORNL REDC. The isotopic composition of this material was measured via mass spectrometry. For major constituents, mass spectrometry results typically have uncertainties on the order of 1%. For the curium feedstock material, this level of uncertainty would apply to Cm-244 and Cm-246. For trace constituents, mass spectrometry measurements are commonly reported with uncertainties of 10% if the measurement is significantly above the instrument detection limit (IDL). This level of uncertainty would apply to the other trace constituents: Cm-245, Cm-247, and Cm-248.

The ORNL REDC curium feedstock material was analyzed via mass spectrometry on at least two occasions: February 20, 1979, and January 8, 1986 (Reference 3). Table 7-2 shows a comparison of the results of these measurements decayed corrected to a common date (January 8, 1986).

Table 7-2. Comparison of Mass Spectrometry Results Decay Corrected to a Common Date

Radionuclide	Results from 2/20/1979 Decay Corrected to 1/8/1986 (at. %)	Results from 1/8/1986 (at. %)	Average	Standard Deviation	Relative Percent Standard Deviation
Cm-244	91.01%	91.14%	91.08%	0.046%	0.05%
Cm-245	1.39%	1.36%	1.37%	0.010%	0.73%
Cm-246	7.38%	7.31%	7.35%	0.026%	0.36%
Cm-247	0.14%	0.12%	0.13%	0.007%	5.19%
Cm-248	0.08%	0.07%	0.07%	0.003%	4.14%

These results are bounded by the above typical mass spectrometry uncertainties. For this analysis, the uncertainty in the Cm-244 and Cm-246 mass spectrometry results is taken to be 1%, whereas a 10% uncertainty is assumed for the other curium isotopes.

Note that for Cm-244, the uncertainty in the mass spectrometry measurement is part of the overall manufacturer's uncertainty and is not combined in quadrature with the manufacturer's uncertainty.

7.3 Uncertainty in Other Radionuclide Impurity Levels

The impurity levels for other radionuclides are taken from samples of eight batches of purified Cm-244. Table 7-3 shows the standard deviations and relative errors in the results for the eight batches.

Table 7-3. Impurity Levels in Cm-244 Materials

Radionuclide	Pu-238	Pu-239	Pu-240	Pu-242	Am-241	Am-243	Cs-137
Average (g/g of Cm-244)	4.35E-05	0.00E+00	2.09E-02	0.00E+00	2.35E-04	1.97E-02	1.84E-07
Std. Dev. (g/g of Cm-244)	4.30E-05		1.52E-02		1.91E-04	3.79E-03	1.28E-07
RSD	98.8%		72.6%		81.5%	19.3%	69.5%

It is assumed that 10 sources in each drum come from the same batch and that each drum contains at least 50 sources of approximately equal size. As a result, each drum has material from at least five batches. The uncertainty is therefore estimated to be the above standard deviation values divided by the square root of five.

Plutonium-239 and Pu-242 were not detected in the batch samples. These isotopes are principally produced from the decay of Am-243 and Cm-246, respectively. Therefore, the uncertainties in the Pu-239 and Pu-242 impurity levels are taken to be the same as for Am-243 and Cm-246.

Plutonium-240 exists as an initial impurity in the purified curium sent to the manufacturer and as a decay product of Cm-244. The uncertainty in the drum Pu-240 inventory due to the initial impurity level is $\frac{0.716}{\sqrt{5}} = 32.5\%$ of this contribution to the inventory. The uncertainty in the drum Pu-240 inventory due to the decay of Cm-244 is equal to the uncertainty in the Cm-244 inventory in the drum, which is just the manufacturer's uncertainty of 0.71%. This uncertainty applies to the fraction of the Pu-240 inventory resulting from the decay of Cm-244. The total Pu-240 uncertainty is the above two contributors, combined in quadrature, divided by the total Pu-240 inventory (depending on the length of the decay period). The calculation for a 10-year decay period is shown in Table 7-4.

Table 7-4. Example of Total Pu-240 Uncertainty from Decay and Initial Inventory

Pu-240 Values	g/g of Cm-244
Initial Pu-240 Impurity	2.09E-02
Pu-240 from Decay	3.13E-01
Total Pu-240	3.34E-01
Uncertainty in Initial Value	6.79E-03
Uncertainty in Decay Value	2.21E-03
Total Uncertainty	7.14E-03
Percent Uncertainty	2.1%

Table 7-5 shows the Pu-240 uncertainty as a function of the decay period. The bulk of the recovered Cm-244 sources are far older than 10 years. Therefore, the Pu-240 uncertainty for 10-year-old sources provides an upper bound and will be assumed for all drums, regardless of the actual age of the material in the drums.

Table 7-5. Pu-240 Uncertainty as a Function of Decay Period

Decay Period (years)	Pu-240 Uncertainty
5	3.6%
10	2.1%
15	1.7%
20	1.4%
25	1.3%
30	1.2%
35	1.1%
40	1.1%

7.4 Uncertainty in the Decay Time from Last Purification to the Shipping Date

Plutonium-239 and Pu-242 are essentially produced only via the decay of the relatively long-lived Am-243 and Cm-246, respectively. In general, a curium batch is purified shortly before it is shipped to the manufacturer. Decay is calculated from the date of manufacture to the shipping date. This calculation implicitly assumes that the date of manufacture is close to the date of the last purification. The uncertainty in the decay time is assumed to be 1 year. It is further assumed that the sources are, on average, more than 10 years old. Therefore, the uncertainty in the decay time is <10%. An upper-bound uncertainty of 10% is therefore applied to the Pu-239 and Pu-242 impurities at the time of shipping.

7.5 Total Uncertainty

The total uncertainty is calculated by combining the above in quadrature. As will be discussed in the next section, the manufacturer's uncertainty is common to all of the radionuclides and is treated separately. Therefore, the combination in quadrature is performed in two steps:

1. all uncertainties other than the manufacturer's uncertainty are combined and
2. the manufacturer's uncertainty is included to determine the total uncertainty.

Table 7-6 summarizes the total uncertainties, excluding the manufacturing uncertainty (Reference 144).

Table 7-6. Summary of the Radionuclide Uncertainties, Excluding the Manufacturing Uncertainty

Radionuclide	Uncertainty in Manufacturer's Stated Source Size	Uncertainty in Curium Isotopics	Uncertainty in Drum Impurity Levels from Sample Data	Uncertainty in Decay Time from Last Purification to Shipping Date	Total Uncertainty Excluding Mfg. Uncertainty	Total Uncertainty
Pu-238	0.71%		44.2%		44.2%	44.2%
Pu-239	0.71%		8.6%	10.0%	13.2%	13.2%
Pu-240	0.71%		2.1%		2.1%	2.3%
Pu-242	0.71%	1.0%		10.0%	10.0%	10.1%
Am-241	0.71%		36.5%		36.5%	36.5%
Am-243	0.71%		8.6%		8.6%	8.6%
Cm-244	0.71%	.			0.0%	0.7%
Cm-245	0.71%	10.0%			10.0%	10.0%
Cm-246	0.71%	1.0%			1.0%	1.2%
Cm-247	0.71%	10.0%			10.0%	10.0%
Cm-248	0.71%	10.0%			10.0%	10.0%
Sr-90	0.71%		31.1%		31.1%	31.1%
Cs-137	0.71%		31.1%		31.1%	31.1%

7.6 Uncertainty for Derived Parameters

The derived parameters TRU, FGE, PE-Ci, decay heat, and total activity are summations made over the individual radionuclides multiplied by the appropriate weighting factors. Elements of the uncertainty analysis apply to multiple radionuclides, e.g., the manufacturer's uncertainty, the uncertainty in the total uranium, and the uncertainty in the age of the sampled material. These elements will introduce statistical dependencies in deriving the uncertainty of the derived parameter. However, for these Cm-244 sources, the derived parameters are dominated by the Cm-244. This dependency is explicitly considered. The uncertainty in the derived parameters is therefore calculated by combining in quadrature the independent sources of uncertainty for each radionuclide, again multiplied by the appropriate weighting factors. The manufacturer's uncertainty is then combined in quadrature with the independent sources of uncertainty, resulting in the total derived parameter uncertainties, as shown in Table 7-7.

Table 7-7. Example Uncertainties for Derived Parameters

Derived Parameter	Total Uncertainty
TRU	45.3%
FGE	0.71%
PE-Ci	44.0%
Decay Heat	46.8%
Total Activity	45.3%

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