Recovering New Type of Sources by Off-Site Source Recovery Project for WIPP Disposal-16604

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INTRODUCTION TO OSRP ACTIVITIES

The Off-Site Source recovery Project (OSRP) is a unique project managed at Los Alamos National Laboratory (LANL) which recovers, packages, transports and qualifies for disposal, disused sealed sources collected from the U.S. and worldwide. OSRP is operating within the National Nuclear Security Administration (NNSA) Office of Global Material Security which sets the yearly milestones for source recoveries. As of November 2015 more than 35,000 sources were recovered by OSRP from over 1200 locations. The majority of the sources recovered under this project contain the transuranic (TRU) isotopes Am-241, Pu-238 and Pu-239. In 2004, OSRP started to recover Cm-244 sources and in 2006, Cf-252 sources. Currently, the inventory of sources recovered containing these two additional radionuclides, reached over 1100 sources. The analysis done on the transuranic content of the containers packaged with these isotopes showed their eligibility for WIPP disposal.

In 2015, OSRP obtained all the necessary approvals for the defense determination requests for sources containing the Cm-244 and Cf-252 isotopes. This approval opened the avenue for WIPP disposal of these sources. Current efforts are concentrating on: waste stream changes for incorporating these isotopes, software validation for characterizing the isotopes under WIPP compliance and packaging policies, and drum transportation for meeting all the Department of Transportation (DOT) and WIPP acceptance requirements.

TYPES OF PACKAGING USED FOR SOURCE DISPOSAL TO WIPP FOR THE NEW ISOTOPES

OSRP uses two types of eligible packaging configurations for Cm-244 and Cf-252 disposal to WIPP: an S-100 drum and a 12”Pipe Overpack (POC) drum. Both types of packaging are WIPP accepted configurations for Contact Handled (CH) waste, which offer different advantages and disadvantages when used in the case of the two isotopes. The S-100 is a poly-shielded type A, 55 gallon drum, containing a 6”diameter POC. The 12”POC drum is a type A, 55 gallon drum without the internal poly-shield and with a pipe component of 12” diameter. The S-100 drum presents the advantage of poly-shielding which is very important for neutron sources, such as the Cf-252, or Cm-244Be, at the expense of reduced internal volume. The

12”POC drum presents a larger pipe component accommodating many more sources, like Cm-244 alpha emitters or shielded Cf-252, for which the CH dose rate is not an issue.

All the sources prepared for disposal to WIPP are required to be special form, as defined in 49 CFR 173.403. When OSRP recovers normal form sources, these can be packaged in a field closable DOT certified Special Form Capsule (SFC) model II or model III, thus becoming special form. These capsules are then packaged in the type-A containers. The difference between the two types of packaging is that the 12”POC can hold more sources, or a mix of sources and capsules at the expense of less shielding. This becomes important when the TRU content criteria of 100nCi/g of TRU waste has to be met for the drums that are packaged for WIPP disposal [1].

QUALIFYING Cm-244 AND Cf-252 FOR WIPP DISPOSAL

The Curium used in the production of sealed sources in the U.S. is produced at Oak Ridge National Laboratory (ORNL). By itself, Cm-244 is not a transuranic isotope by WIPP definition, due to its short half-life of 18.1 years. However, it qualifies as transuranic due to the Pu-240 daughter in-growth in the alpha decay chain. Pu-240 has a half-life of 6563 years. The composition of the Curium batch material used in sealed source manufacturing has about 86-91% Cm-244. In U.S., most of these sources were produced by private source manufacturers, e.g. QSA Global, Eckert Ziegler (IPL), and Frontier Technology. A smaller number were produced by national labs, targeted towards specific research applications. In some of the sources, Cm-244 was mixed with Beryllium, Be, creating an (α, n) neutron source with various neutron output.

The Californium used in the sealed production in the U.S. is produced at ORNL. This isotope has a 2.65 years half-life and it decays by both α emission and spontaneous fission. Some of the other Californium isotopes present in the production batch material are Cf-249, Cf-251 as well as Cm-248 which is the α decay product from Cf-252. These contribute to the transuranic content of the Cf-252 sources. The average composition of the Californium batch material used in the sealed source manufacturing has about 81-83% of Cf-252. This isotope makes a great neutron source with emissions in the range of 10^9 to 10^{10}neutrons/s /Ci, [2] with the biggest advantage being its small mass and volume while providing a large neutron yield.

RESULTS ON DOSE RATES MEASUREMENT FOR Cf-252 DRUMS

Dose rates were measured on several drums packaged with Cm-244, Cm-244Be and Cf-252 sources. The neutron dose rates were measured with Smart Neutron Rem Detector® (SNRD) and the gamma dose rates were measured with an Eberline Model E-600 connected to a SHP Eberline probe. The studied sources were packaged in the two drum configurations discussed earlier, the S-100 and the 12”POC. At the same time, the amount of transuranic material was calculated for the packaged sources, using the characterization software developed for WIPP disposal of the 2 isotopes. The decayed values for the Cm-244 and Cf-252 concentrations were used as input into a built-in model with MCNP code [3] for the S-100 and 12”POC drums to verify if the modeled results for dose rate compare with the real time measurements. The correlation that provides the contact estimated neutron dose rate for a known Cf-252 activity loading for an S-100 drum, is presented in equation (1):

$$y = 382.75\,x \qquad (1)$$

where, y = drum contact neutron dose rate, mrem/h

382.75 = correlation factor, mrem/h/Ci of Cf-252 decayed and

x= decayed activity of Cf-252, Ci.

A drum containing a Cf-252 source of 1.7 Ci, produced at ORNL in 1981, was used to verify the correlation developed with MCNP between the current Cf-252 drum loading and the neutron dose rate. The neutron dose rate is the result of the spontaneous fission of the Cf-252 in the S-100 drums. Table I shows the neutron and gamma dose rate measurements on contact for a Cf-252 source packaged in an S-100 drum. The model for calculating the gamma dose rates has not been finalized at this time.

Table I. Contact Dose Rate for Cf-252 Source for an S-100 Drum

Type of Dose	**Measured Dose Rate, mrem/h**	**Calculated Dose Rate, mrem/h Cf-252**	**Calculated Dose Rate, mrem/h Cf-250**	**Calculated Total Dose Rate, mrem/h**
Neutron	0.57 ± 0.40	0.08	0.08	0.16
Gamma	1.11 ± 0.41	Not Available	Not Available	Not Available

The measured neutron dose rate results came out consistently higher than the modeled dose rate for Cf-252. The measured neutron values were only twice than the background, therefore incorporating the errors into the measurement needed to be addressed. The total uncertainty for contact neutron measurements for the Cf-252 source in the S-100 drum was calculated to be about 70%. The Cf-252 neutron measurement error calculation included the errors due to the measurements, the distance, the calibration of the instrument and the source activity uncertainty.

In equation (1) the correlation developed between the Cf-252 drum loading and the neutron dose rate takes into account only the neutron contribution from the Cf-252. The correlation had to be corrected in order to incorporate the presence of Cf-250 in the original material which has a contribution to the neutron production via spontaneous fission. Table II presents the nuclear information for Californium isotopes [4], [5] and the Average Mass Fraction for the material used in source production.

Table II. Californium Isotopes Nuclear Data and Sealed Source Batch Composition

Nuclide	Half-Life	Average Mass Fraction (%)	Spontaneous Fission (SF) Branching Fraction	Average Number of Neutrons per SF	Total Neutron Emission Rate (n/g/s)
Cf-249	351 y	5.57	$5.2x10^{-7}$	3.4	$6.34x10^{3}$
Cf-250	13.1 y	9.98	0.077	3.53	$1.12x10^{10}$
Cf-251	898 y	3.11	$9.0x10^{-6}$	3.7	$1.96x10^{6}$
Cf-252	2.65 y	81.26	0.03096	3.77	$2.31x10^{12}$
Cf-253	17.81 d	0.06	Not Reported	Not Reported	$8.41x10^{4}$
Cf-254	60.5 d	0.02	0.31	3.93	$1.20x10^{15}$

In a new source, the spontaneous fission emission rate is dominated by the Cf-252. The influence of Cf-250 on the neutron measurement increases with time as the Cf-252 concentration decreases due to its short half-life. The contribution of Cf-250 to the total neutron emission rate for a Cf-252 source increases to 50% for a 30y old source. In this case, the neutron yield from Cf-250 becomes the same order of magnitude as the decayed Cf-252, and is of the order of 10^8 neutrons/s. The ratio of total neutron emission rate of Cf-252 to Cf-250 was used to correct for the additional neutrons calculated for the S-100, on contact. For the 34 year old studied source, a Cf-252:Cf-250=1:1 neutron emission rate ratio was used for neutron calculations. Table I presents the calculated results for neutron dose rate from Cf-250 for the source used in the measurements.

The modeled neutron dose rate results for a 34 year old source packaged in an S-100 drum is within the 2 standard deviation applied to the measured values. Given the large uncertainty in the measured Cf-252 neutron contact dose rate, and the need to fully validate the MCNP model, a stronger Cf-252 neutron source and a lower background will be used in the future to take measurements, and correlate the neutron dose rate for the packaging of Cf-252 source in the S-100 drum configuration.

The measured gamma dose rate in the S-100 for the Cf-252 source is primarily due to the (n, γ) reaction with the poly material. The correlation for the gamma dose rate for Cf-252 has not been fully validated.

A similar correlation as in equation (1) was developed for 12”POC. The same 34 year old Cf-252 source was packaged in a 12” POC. The MCNP developed correlation that provides the contact estimated neutron dose rate for a known Cf-252 activity loading in a 12”POC drum, is presented in equation (2):

$$y = 23{,}000\, x \qquad (2)$$

where, y = drum contact neutron dose rate, mrem/h

23,000= correlation factor, mrem/h/Ci of Cf-252 decayed and

x= decayed activity of Cf-252, Ci.

The contact neutron and gamma dose rate measurements for a Cf-252 source packaged in a 12” POC and the modeled neutron dose rate are presented in Table III:

Table III. Contact Dose Rate for Cf-252 Source for a 12”POC Drum

Type of Dose	Measured Dose Rate, mrem/h	Calculated Dose Rate, mrem/h Cf-252	Calculated Dose Rate, mrem/h Cf-250	Calculated Total Dose Rate, mrem/h
Neutron	15.4 ± 5.7[a]	4.9	4.9	9.8
Gamma	2.1 ± 0.8[a]	Not Available	Not Available	Not Available

[a] *37% total uncertainty for contact dose*

The total uncertainty for neutron contact measurements for the Cf-252 source in the 12”POC drum was calculated to be of 37%. When taking into account the standard deviation for the measured values, the measured and calculated neutron dose rate based on equation (2) are in reasonable agreement.

RESULTS ON DOSE RATES MEASUREMENT FOR Cm-244 DRUMS

The contact neutron and gamma dose rates for an S-100 drum were measured for a Cm-244Be source. The source was packaged in a SFC model II, with an original concentration of 57.5 Ci of Cm-244 in 1970. At the time of these measurements, it had decayed to about 10.4 Ci of Cm-244. This source produces about 10^4 nCi/g capsule and it qualifies for WIPP disposal. The Cm-244Be source produced similar dose rate results equivalent to Am-241Be sources. Previous data for Am-241Be sources in S-100 drums showed an approximately 1:1 ratio of neutron to gamma dose rates. Table IV shows the comparison between dose rates produced by Am-241Be and by Cm-244 Be sources measured in the S-100 container.

Table IV. Measured Contact Dose Rates for Cm-244Be and Am-241Be Sources for an S-100 Drum

Isotope	Contact Neutron Dose(mrem/h/Ci)	Contact Gamma Dose(mrem/h/Ci)	Total Dose Rate(mrem/h/Ci)
Am-241Be	1.18	1.03	2.23
Cm-244Be	1.14 ± 0.42[a]	0.92 ± 0.34[a]	2.06 ± 0.54[a]

[a] *37% total uncertainty for contact dose*

The evaluation of the data in Table IV shows the similarity between contact dose rates produced by a Cm-244Be source and an Am-241Be source. For these types of sources one can expect a total neutron and gamma dose rate close to 2 mrem/h/Ci of decayed Cm-244 and a ratio of 1:1, of neutron to gamma dose rates.

A large Cm-244 source packaged in an S-100 drum was used to measure the drum neutron and gamma dose rates on contact. The source had 71.5 Ci of Cm-244 in 2015, at the time of these measurements. This source alone produces 10^5 nCi/g capsule, well over the 100nCi/g necessary for WIPP disposal. The MCNP developed correlation that provides the contact estimated neutron dose rate for a known Cm-244 activity loading in an S-100 drum, is presented in equation (3):

$$y = 0.015\, x \qquad (3)$$

where, y = drum contact neutron dose rate, mrem/h

0.015= correlation factor, mrem/h/Ci of Cm-244 decayed and

x= decayed activity of Cm-244, Ci.

The contact neutron and gamma dose rate measurements for Cm-244 source packaged in an S-100 drum as well as the modeled neutron dose rate are presented in Table IV:

Table IV. Contact Dose Rate for Cm-244 Source for an S-100 Drum

Type of Dose	Measured Dose Rate, mrem/h	Calculated Dose Rate, mrem/h
Neutron	1.30 ± 0.48[a]	1.07
Gamma	4.10 ± 1.52[a]	Not Available

[a] *37% total uncertainty for contact dose*

The measured and calculated neutron dose rates for the Cm-244 source packaged in an S-100 drum agree reasonably well. The model for calculating the gamma dose rates is has not been finalized at this time.

CONCLUSIONS AND FUTURE WORK

Measured neutron dose rates for Cm-244 packaged in S-100 agree reasonably well with calculated dose rates based on the models developed with MCNP. Future work will include validating the model for Cm-244 packaged in 12”POC. A 1:1 ratio of neutron to gamma is expected for Cm-244Be dose rates packaged in S-100 drums.

For Cf-252 packaged in S-100 and 12”POC, the contribution of Cf-250 to the total emission neutron rate had to be taken into account due to the increasing role of Cf-250 as sources age. The uncertainty in the measurement of Cf-252 neutron dose rate for S-100 drum was quite high due to a high neutron background. A larger Cf-252 source needs to be used for measurements in the future, in order to fully validate the model developed. A better agreement for measured versus calculated rate was obtained for a Cf-252 packaged in 12”POC.

For the WIPP disposal of these sources, OSRP will use the best packaging configuration for these isotopes in order to minimize the dose rate and maximize the TRU content. The S-100 packaging will be used for Cf-252 and Cm-244Be sources, when neutron dose reduction by the poly-shielding is important. Neutron dose rates will be estimated using the new MCNP correlations.

REFERENCES

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