Approach for the Isotopic Characterization of Irradiated Start-Up Sources for Disposal at WIPP - 15674

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ABSTRACT

The National Nuclear Security Administration, Global Threat Reduction Initiative (GTRI)/ Off-site Source Recovery Program (OSRP) requires an approved final disposition pathway for sealed irradiated startup sources with uncertain irradiation histories. To create this path a reliable method is required to determine yields of transmutation, fission and activation products contained within the source at the time of disposal. Because of the uncertain histories of these sources, a number of likely histories are modeled with the code ORIGEN in order to establish the bounds of the radioisotope yields. The ORIGEN simulations are confirmed with gamma spectroscopy, specifically examining the activities of several fission and transmutation isotopes. These calculations, coupled with other previously established characterization techniques for transuranic neutron sources, will be used to establish a waste profile that is in compliance with Waste Isolation Pilot Plant (WIPP) waste acceptance criteria.

INTRODUCTION

The mission of the OSRP is to remove from the public domain unwanted, excess, and abandoned radioactive sealed sources that pose a potential threat to health or national security [1]. Initially the OSRP was mandated to collect transuranic Greater Than Class C (GTCC) low-level radioactive waste. After 2001 the mission expanded to include the collection of beta and gamma emitting sources of concern to the NNSA in addition to the GTCC waste. Characterization of the sources collected by the OSRP has used Acceptable Knowledge (AK) [2]. Typically, AK documentation consists of source markings, or manufacturing documents certifying the initial composition and manufacturing dates of the collected sources. In cases where the sources have not been exposed to activating radiation, this documentation adjusted with an appropriate decay calculation is sufficient to determine the isotopic composition of the sources at the time of their disposal [3]. However, over the life of the project a number of sources have been made available for collection, with a history of exposure to activating radiation during their lifetimes. These sources do not have an approved characterization methodology and are being held at OSRP facilities or are awaiting collection from the licensee. A subset of these sources consists of AmBe reactor startup sources which are in storage at reactor sites awaiting recovery and disposal. To determine the isotopic composition of this family of sources an ORIGEN [4] simulation based upon a likely burn will be developed and then validated using observed gamma radiation emitted from the source.

METHODS

In order to derive the source isotopic content after exposure to activating radiation it is required to know the initial composition of the startup source and the power history of the reactor for the time when the source was in-core. The approximate removal date from the core is also necessary to calculate the post-irradiation decay, giving the composition at the time of waste disposal.

A typical startup source consists of a mix of americium oxide physically blended with Beryllium and doubly encapsulated in a stainless steel capsule. The mechanism for production of neutrons from this source consists of the (α, η) reaction where alpha particle emission from the decay of the Am-241 are then captured by the Be-9, releasing a neutron and a C-12 atom. The initial composition of the source material is well understood and well documented via the manufacturing documents, and other AK documentation. Exposure of this initial source results in transmutation of isotopes as well as the production of fission isotopes. The activation of the small fraction of naturally occurring Fe-58 (0.282%) in stainless steel produces Co-60 via the decay of Fe-59 and the subsequent (η, ϒ) absorption of Co-59 to produce C0-60. Regrettably, the exposure history of the sources during their service is typically unavailable as may be the time of their removal from the reactor core. However, gamma spectroscopy and neutron counting are available for analysis of the radiation emitted from the source at the time of recovery.

The SCALE 6.1 software package [4] is used to simulate the evolution of the source composition during its burn history, and during the decay after the removal from the core. Reactor configuration for input into the code was based on a standard fuel design and configuration using a 16 by 16 fuel array unbiased by the presence of the source. Reactor power levels were also taken as an average of the power generated by the reactor when the source was present in the core. These parameters were entered into the Automatic Rapid Processing (ARP) code in SCALE to generate effective cross sections for use in the ORIGEN code. The large amounts of fuel relative to the startup source materials resulted in the fuel reaction products masking the source products in the simulation, necessitating running simulations without the fuel present. The core neutron flux was then calculated from the power using ORIGEN with no source present and passed into the source only simulations. It is assumed that the flux influence caused by the source is negligible given the amount of fuel in the core and has no impact on the in-core flux modeling.

Because the exact burn history of the source in the core is often unavailable, a number of different histories were generated with ORIGEN to account for variations in possible burn scenarios. A two month burn history was included in the modeled power history to account for cases where the source was left in for startup and removed once initial startup and testing was complete. An average energy and low energy cases were modeled with this time window for the cases when the reactor was running at low power for the startup or if the reactor was running at normal power for most of the time. A one year long simulation was also generated for cases where the source was left in the core for the initial burn cycle until it was removed during fuel rearrangement. A continuous burn process and an interrupted burn process were also modeled

for cases where there were significant downtimes occurred during the burn cycle. For all cases a decay time of 12 years after removal from the core was modeled with isotopic yields and activities recorded at time of removal and then annually until the end of the 12 years.

DISCUSSION

To accurately predict the composition of the source it is necessary to develop a method to determine the irradiation history. Evaluation of the simulated histories shows that the parameter with the most significant impact on the yield is the time in core (i.e. total flux). Lower flux rates and interruptions in the burn process caused small changes to the isotopic compositions by reducing the probability that the short lived isotopes would react with the reactor neutrons before they decayed away. The effects of these interruptions on a sample of WIPP reportable isotopes [5] can be seen in Figure 1.

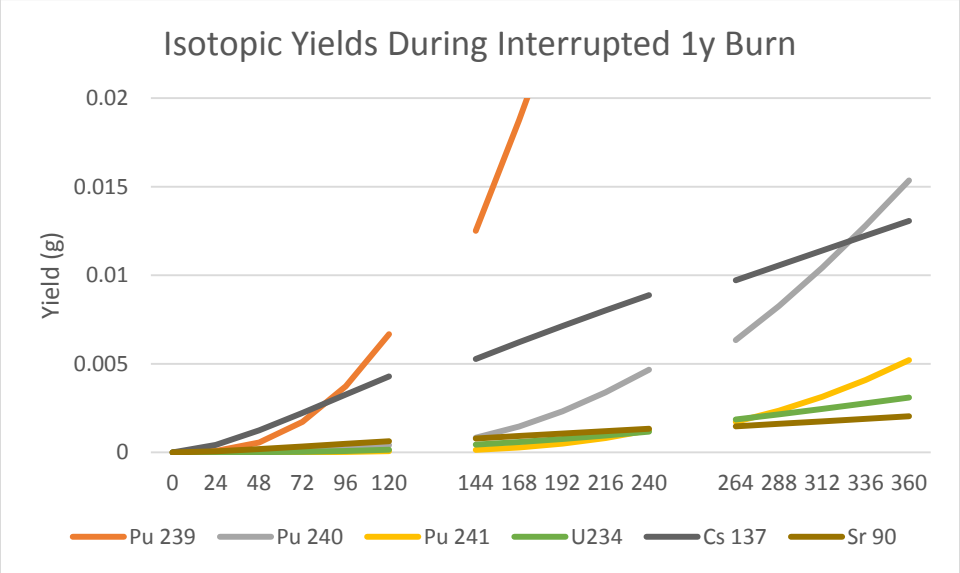

Figure 1. Isotopic yields for selected isotopes during an interrupted burn.

The in-growth rate as a function of total flux varies between isotopes in the source. The variation can be seen most clearly in the growth rates of the fission products and the transmutation products. Examples of this are Cs-137 and Cm-243 shown in Figure 2.

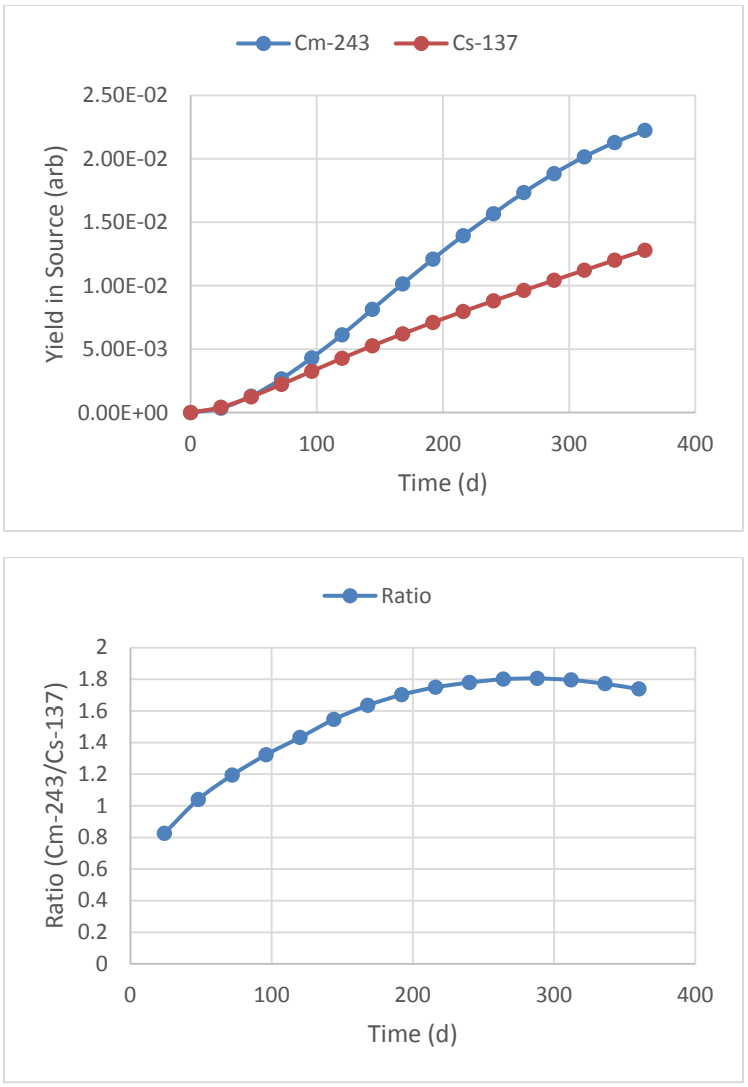

Figure 2. (Top) In-growth of Cm-243 and Cs-137 during the course of a 1-year uninterrupted time in core. (Bottom) Ratios of Cm-243 and Cs-137 during the same period.

Because the growth rates as a function of flux are different between the isotopes it is possible to determine the integral flux the source was exposed to by examining the ratios of isotopes contained in the source. Gamma spectroscopy can be used to quantify the activities of radioisotopes observed in the source and thus generate their ratios. The calculation of the isotope ratios after time of removal is complicated by the fact that if the source has been removed from exposure for an unknown period of time before the measurement the initial ratios of isotopes with different half-lives cannot be determined. However, this difficulty can be circumvented by choosing isotopes with similar half-lives for analysis. For the purposes of determining the amount of flux an AmBe source was exposed to, we require isotopes that can be observed though gamma spectroscopy, have similar half-lives and have half-lives long enough that they will exist

in observable quantities for years after removal from the core. The most prominent gamma lines predicted to be observable are shown in Figure 3.

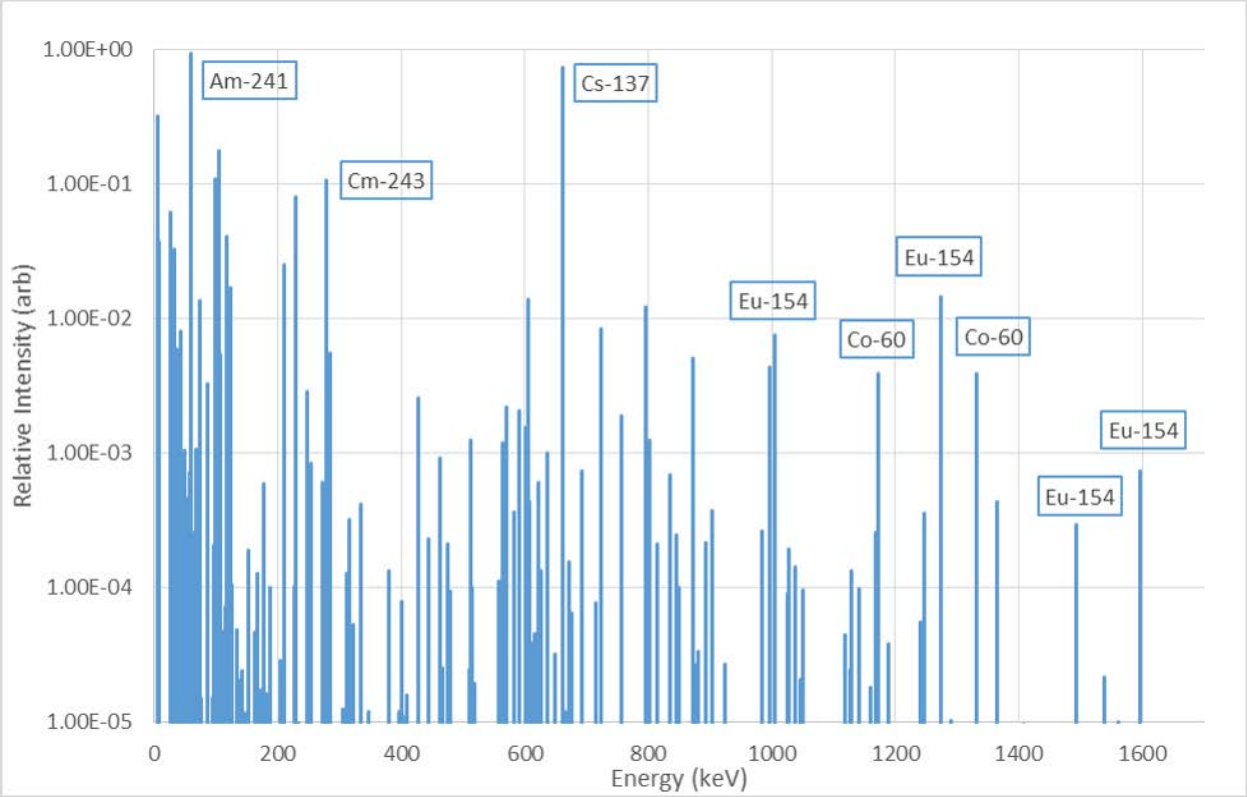

Figure 3. Calculated gamma spectrum for AmBe source irradiated for 1-year in 400 MW reactor core and then removed and allowed to decay for 12 years. Gamma lines of possible use for determination of total flux are indicated.

From the discussion above, Cs-137 and Cm-243 are the best candidates for determination of the total flux. They are both observable, with significantly differing (non-parallel) in-growth rates and their half-lives are near identical (29.1y for Cm-243 and 30.08y for Cs-137). Because the half-lives are similar the ratio between Cs-137 and Cm-243 will remain nearly fixed after removal from the core. This will allow for a calculation of total flux that is independent of time spent out of core. Comparing Cs-137 or Cm-243 to an isotope with a different half-life, for example Eu-154 will then allow determination of the time removed from the core defining the parameters necessary for the simulation of the source composition.

The Pu-238 activity of the source increases the alpha activity by a factor of 2.5 after 1y of exposure and 12 y of decay, seen in TABLE I. Since Pu-238 is an alpha emitter the increase in alpha radiation will be seen in an increase in the neutron count rate emitted by the source proportionate to the increase in Pu-238 activity. This second measurement can then be used to confirm the calculations of isotope yield based upon the Cm-243/Cs-137 ratio. An additional confirmation may be possible by measurement of the Co-60 content of the source caused by transmutation of the Fe in the stainless steel cladding and comparing it to calculations based on the known dimensions of the source cladding.

TABLE I. Activity of primary alpha emitters before exposure, 1y and 12y after removal from core.

Initial Alpha		1y Alpha		12y Alpha	
Isotope	Activity (Ci)	Isotope	Activity (Ci)	Isotope	Activity (Ci)
Am-241	10.41	Cm-242	388.70	Pu-238	20.46
		Pu-238	20.31	Am-241	2.56
		Am-241	2.60	Cm-244	1.53
		Cm-244	2.33	Cm-243	0.76
		Cm-243	0.99	Cm-242	0.16
Total	10.41	Total	414.93	Total	25.47

Uncertainty analysis based upon the limits of measurement accuracy and the sensitivity of the simulations to assumptions about burn conditions will provide upper and lower bounds for the isotopic composition of the irradiated sources. This uncertainty calculation will be used to ensure that the determination of the source composition falls within the characterization requirements of WIPP [6].

CONCLUSIONS

The successful characterization of irradiated startup sources can be accomplished by the combined use of computer modeling with the spectroscopic assay of individual sources. The determination of the isotopic content of the sources and the ratios of isotopes with similar half-lives should allow for the determination of total neutron exposure via comparison to modeled data. This information further corrected for out of core decay will allow for determination of the detailed isotopic content of the irradiated source and permit final disposition of this transuranic material to WIPP.

REFERENCES

[1] US Department of Energy, *"Plan to Ensure Continued Recovery and Storage of Greater-Than-Class C Low-Level Radioactive Sealed Sources that Pose a Security Threat Until a Permanent Disposal Facility is Available"*, Report to US Congress, (2005)

[2] Los Alamos National Laboratory, CCP-AK-LANL-008, Revision 9, *"CCP Acceptable Knowledge Summary Report for LANL OSRP Sealed Sources"*, May, 2012

[3] Vance and Associates, *"Development of Radionuclide Distribution and Uncertainties in Am-241 Sealed Source Material"*, OSR-TD-012; 2005

[4] *SCALE: A Comprehensive Modeling and Simulation Suite for Nuclear Safety Analysis and Design*, ORNL/TM-2005/39, Version 6.1, Oak Ridge National Laboratory, Oak Ridge, Tennessee, June 2011

[5] US Department of Energy, *"Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant"*, Revision 7.2, June, 2011

[6] B. T. Rearden, M. L. Williams, M. A. Jessee, D. E. Mueller, and D. A. Wiarda, "Sensitivity and Uncertainty Analysis Capabilities and Data in SCALE," *Nucl. Technol*. **174**(2), 236-288, May 2011.