

UCRL-JRNL-215027



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Analyzing Nuclear Fuel Cycles from Isotopic Ratios of Waste Products Applicable to Measurement by Accelerator Mass Spectrometry

S. R. Biegalski, S. M. Whitney, B.A. Buchholz

September 6, 2005

Nuclear Science and Engineering

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

Analyzing Nuclear Fuel Cycles from Isotopic Ratios of Waste Products Applicable to Measurement by Accelerator Mass Spectrometry

Scott M. Whitney¹, Steven Biegalski^{1*}, Bruce Buchholz²

1. *The University of Texas at Austin, PRC-Nucl Engr Teach Lab, 1 University Station
Stop R9000, Austin, TX, 78741*

2. *Lawrence Livermore National Laboratory, CAMS, Livermore, CA 94551*

*biegalski@mail.utexas.edu

Abstract

An extensive study was conducted to determine isotopic ratios of nuclides in spent fuel that may be utilized to reveal historical characteristics of a nuclear reactor cycle. This forensic information is important to determine the origin of unknown nuclear waste. The distribution of isotopes in waste products provides information about a nuclear fuel cycle, even when the isotopes of uranium and plutonium are removed through chemical processing. Several different reactor cycles of the PWR, BWR, CANDU, and LMFBR were simulated for this work with the ORIGEN-ARP and ORIGEN 2.2 codes. The spent fuel nuclide concentrations of these reactors were analyzed to find the most informative isotopic ratios indicative of irradiation cycle length and reactor design. Special focus was given to long-lived and stable fission products that would be present many years after their creation. For such nuclides, mass spectrometry analysis methods often have better detection limits than classic gamma-ray spectroscopy. The isotopic ratios

$^{151}\text{Sm}/^{146}\text{Sm}$, $^{149}\text{Sm}/^{146}\text{Sm}$, and $^{244}\text{Cm}/^{246}\text{Cm}$ were found to be good indicators of fuel

cycle length and are well suited for analysis by accelerator mass spectrometry.

Introduction

The production of fission products and transuranics in a reactor system is dependent upon specific features and physical parameters of the reactor system such as reactor fuel type, moderator, and overall design. Some of these features may be determined by the isotopic profiles of waste from a reactor. Analysis of the fission products may reveal history of the fuel such as time spent in the reactor core, and in some cases, even the type of reactor from which the spent fuel originates. Since ratios of important nuclides may vary depending on the length of a reactor cycle, then assessing the length of reactor cycles may be quite important in some situations.

Often times key fuel cycle indicators such as plutonium and uranium are not available (*e.g.*, after reprocessing). Considering such a situation, this work establishes which long-lived isotopes are most illustrative of fuel cycle history. It is shown that analysis for specific isotopic ratios contained in the waste after spent fuel reprocessing is indicative of the length of the fuel cycle as well as reactor type when sensitive detecting techniques are utilized. It is important to note that isotopic ratios are utilized in contrast to ratios of different elements since isotopic ratios are least affected by chemical fractionation.

Furthermore, conclusive evidence was found expressing the need for sensitive analytical techniques if the spent fuel is to be accurately analyzed for the entirety of information it can provide. Particular isotopes in the spent fuel have characteristics that prevent accurate analysis via conventional techniques such as gamma-ray spectroscopy. Analysis of samples that are small or diluted via environmental transport further

exacerbates the hardships of analyzing these particular isotopes and necessitates the use of mass spectrometry detection techniques [1].

Accelerator mass spectroscopy (AMS) is one analytical technique with the capabilities of measuring as little as 10^5 atoms [2,3]. AMS is also particularly useful for analysis of isotopes with long half-lives, between 10 and 10^7 years, and low gamma-ray yields. The data presented in this manuscript shows that in many instances radionuclides with low gamma-ray yields are very informative of reactor cycle parameters. AMS requires one to obtain only a very small sample size, on the order of micrograms in some cases, and enables the analysis of isotopes immeasurable by other analytical techniques [2,3].

Most AMS studies with isotopes of interest to the nuclear fuel cycle have examined environmental releases or biological exposures to uranium and plutonium[4], with a few limited forays with ^{63}Ni [5], ^{90}Sr [6], ^{99}Tc [7], and ^{129}I [8]. Our work sorts through the other radionuclides produced in the nuclear fuel cycle to determine which of these radionuclides have the half-life and chemistry suitable for AMS analysis. The isotopes that are most informative of particular parameters of the reactor cycle in which they were produced were determined. A hierarchy of those isotopes that are most useful was produced and the top isotopes were identified. The methods produced for this work identify isotopic ratios that may be used to distinguish commercial power fuel cycles from other fuel cycles.

ORIGEN Modeling

Simulations of several different reactor cycles were performed with the Oak Ridge Isotope Generation and Depletion codes, ORIGEN-ARP and ORIGEN-2.2 [9, 10]. These codes were used to model the reactor cycles of the Pressurized Water Reactor (PWR), Boiling Water Reactor (BWR), Canada Deuterium Uranium Reactor (CANDU), and Liquid Metal Fast Breeder Reactor (LMFBR).

Parameters of each reactor cycle were specified such as power and initial composition. Other parameters of the cycles like irradiation time were varied throughout our study. Simulations were initiated with ORIGEN-ARP to gain a general knowledge of which isotopes to focus on with PWR and BWR cycles. ORIGEN 2.2 was later used to model the PWR, BWR, and also the CANDU and LMFBR.

The focus of this study was to determine which isotopes best differentiate between the variables of reactor cycles. Low and high burnup conditions were modeled to identify which isotopic concentrations differed most. A low burnup or “short” irradiation cycle simulation was approximately one month. The high burnup or “long” irradiation cycle was 540 days (18 months), and is typical of a commercial light water power reactor.

Obtaining a sample immediately after irradiation is unlikely, so each calculation assumes either a “short” or “long” fuel cycle and a decay period of one year. Input decks for the ORIGEN-2.2 code were written to simulate long and short cycles for each reactor design. The input decks were very similar but differed slightly depending on the reactor. The ORIGEN code that was used to model the reactors varied with respect to power, cross-section libraries, and initial compositions. Depending upon the reactor being

simulated, different enrichments of ^{235}U and the absence or presence of initial Plutonium, was specified in the input deck of the code.

The ORIGEN codes are a well substantiated set of codes that have been used for decades in nuclear engineering research. According to U.P. Jenquin and R.J. Guenther [9], the ratio of ORIGEN-2 code calculated values of Uranium and Plutonium isotopes to measured values from actual spent fuel to be in the range of 0.94 to 1.03 for the PWRU cross-section libraries*. A similar study by T. Adachi *et al.* [24] shows other actinide calculations to reveal no significant difference within about 10% of measured values from spent fuel. A combination of noble gas mass spectrometry, reactor modeling and further data analysis corroborated declared fuel burnup to within 4% from a light water reactor [11].

Further studies by Hermann and DeHart have found “reasonable agreement between measured and predicted isotopic concentrations for important actinides,” for BWR spent fuel [12]. A separate study comparing benchmark isotopic samples to the ORIGEN-S codes found that “the code accurately models a broad spectrum of problems to which it is typically applied,” [13]. Since our purposes in this project focus on order of magnitude types of values, any small differences in concentrations will not be significant during any actual analysis.

Methods of Analysis

The primary goal of this study was to determine the isotopic pairs that are most useful for differentiating between cycle length and reactor design. Knowledge of these

* PWRU was the cross-section library used in ORIGEN 2.2 for PWR calculations.

parameters may give key insight into the source of fission products found with unknown origin. Using isotopic pairs rather than a single isotope of an element eliminates the discrepancies in many of the initial conditions such as total amount of initial composition and power. Analysis of isotopic pairs also minimizes defects due to variations in chemical separation efficiencies between different elements.

The isotopic pairs that are most informative for this study are those pairs that show the most dramatic differences in production with respect to the short or long cycle. To quantify this idea, the output of each reactor simulation was analyzed to form a ratio of isotopic pair ratios. This is the ratio of an isotopic pair from the short cycle to the same isotopic pair from the long cycle. This value is the cycle ratio, R_c . The formula for R_c is provided in equation 1.

$$R_c = \frac{\frac{({}^A_Z X)_{short}}{({}^{A-i}_Z X)_{short}}}{\frac{({}^A_Z X)_{long}}{({}^{A-i}_Z X)_{long}}} \quad (1)$$

The values of R_c were calculated for every fission product as well as the mass of each fission product.

Sample Size, M_s

Although the values of R_c are informative about the ability of an isotopic pair to differentiate between whether it was produced in a long or short cycle, it is also important to actually detect the isotope masses in a given sample from a reactor's spent fuel. For this assessment, a detection limit needs to be calculated. The benefit of Accelerator Mass Spectrometry (AMS) is accentuated at this point since AMS is capable of detecting

samples of atoms as low as 10^5 atoms. In an effort to limit uncertainty in analysis, the lower limit of detection was assumed to be 10^9 for the calculations below.

Assuming a uniform distribution of the isotopes throughout the fuel, the sample size needed to obtain 10^9 atoms of the isotope being studied was calculated. Therefore, the minimum mass of unprocessed spent nuclear fuel containing the requisite mass of each of the desired fission or activation products, M_s , was calculated. Since the isotopes are evaluated in the form of isotopic pairs, M_s the pair is limited by the larger value of M_s for the two isotopes. Equation 2 shows the calculation for M_s .

$$M_N = \frac{(10^9 \text{ atoms})(MW)}{N_A}$$

$$\frac{M_s}{M_N} = \frac{M_T}{M_E} \tag{2}$$

$$M_s = M_N \left(\frac{M_T}{M_E} \right).$$

where,

M_T = total mass of spent fuel from reactor cycle in ORIGEN calculation

M_E = total output mass of isotope from simulated reactor cycle in ORIGEN calculation

M_N = mass needed for 10^9 atoms of the isotope

MW =molecular weight in g mol^{-1}

N_A =Avogadro's number.

Values for M_s vary significantly reaching as low as micrograms and as high as 10^{31} grams, a mass five times that of Jupiter. To determine the isotopic pairs that are most useful to us, it was necessary that we consider both the cycle ratio as well as the required minimum sample size, M_s .

Reactor Design Differentiation

Similar to the computation of R_c , the value of R_d was calculated to quantify the ability of an isotopic pair to differentiate between reactor designs. As shown in equation 3, R_d enumerates the ability to determine the type of reactor in which the isotopes were created.

$$R_d = \frac{\left(\frac{{}^A_Z X}{PWR}\right) / \left(\frac{{}^{A-i}_Z X}{PWR}\right)}{\left(\frac{{}^A_Z X}{LMFBR}\right) / \left(\frac{{}^{A-i}_Z X}{LMFBR}\right)} \quad (3)$$

Equation 3 only represents one example of the 6 possibilities of R_d values comparing the PWR, BWR, CANDU, and LMFBR.

Accelerator Mass Spectroscopy (AMS)

Accelerator Mass Spectroscopy is an analytical method with outstanding detection capabilities [2,3]. As previously mentioned, the ability of AMS to detect $\sim 10^5$ - 10^6 atoms enhances the possibilities of gaining a large amount of information from a very small sample. In this section those isotopes that are best suited for AMS analysis are highlighted.

Most detection systems rely on nuclear processes such as gamma emission in order to detect and identify the isotope being observed. Conversely, AMS counts atoms rather than waiting for radioactive decay events. As a result, long half-lives and low gamma yields are not problems for AMS detection limits. Many long-lived isotopes that are produced in the reactor cycle are not suitable for detection via traditional

spectroscopy methods. Analysis of R_c and R_d shows that many of these long-lived isotopes are very informative of the nuclear history.

The gamma counts emitted from every long-lived (i.e. $t_{1/2} > 10$ years) isotope was analyzed to gain an idea of the isotope's possibility for detection. It was assumed that a range of about 100-250 counts in a counting time of 24 hours would be detectable by traditional spectroscopic methods. In these calculations, it was assumed that a detector efficiency of 4% and a counting time of 24 hours. Using equation 4, we approximated the counts for each isotope. Note that count-rate is assumed to be constant over these counts since the half-life is so long.

$$\begin{aligned} R &= A_0 \gamma_E \epsilon_E t_c \\ R &= \lambda N_0 \gamma_E \epsilon_E t_c \end{aligned} \quad (4)$$

$$\lambda = \text{decay constant} = \frac{\ln 2}{t_{1/2}} \quad N_0 = \text{initial number of atoms} = 10^9 \text{ atoms} \quad \gamma_E = \text{Gamma intensity}$$

$$\epsilon_E = \text{detector efficiency} = 0.04 \quad t_c = \text{count time} = 24 \text{ hours}$$

All isotopes that were calculated to have an insufficient number of counts (<250) for normal detection were noted as AMS isotopes. Table 1 lists the isotopes found to be more suitable for AMS than for spectroscopy.

Table 1. Nuclides more suitable for AMS in contrast to gamma-ray spectroscopy.

Nuclide	Nuclide	Nuclide	Nuclide
²⁴¹ Am	¹⁵¹ Eu	²⁰² Pb	¹⁵⁰ Sm
²⁴³ Am	¹⁸² Hf	²¹⁰ Pb	^{121m} Sn
²⁰⁸ Bi	^{166m} Ho	¹⁰⁷ Pd	¹²⁶ Sn
²⁴⁴ Cm	⁸¹ Kr	^{186m} Re	⁹⁰ Sr
²⁴⁵ Cm	⁸⁵ Kr	¹⁴⁶ Sm	⁹⁸ Tc
²⁴⁶ Cm	⁹⁴ Nb	¹⁴⁷ Sm	⁹⁹ Tc
²⁴⁷ Cm	²³⁶ Np	¹⁴⁸ Sm	²²⁹ Th
²⁴⁸ Cm	²³⁷ Np	¹⁴⁹ Sm	²³⁰ Th

The list above makes no claims as to the production of each isotope in a reactor cycle. It just states that the isotopes of table 1 are better suited for AMS analysis in contrast to classical gamma-ray spectroscopy due to their long half-life and/or low gamma activity. Many of these isotopes offer very promising R_c and M_s values when paired with other isotopes.

Results

Spent fuel material allowed to decay for a year or more after a reactor cycle is nearly devoid of many informative short-lived isotopes. In some special cases there is no knowledge of when the reactor cycle ended or how long the sample has been decaying. In this case, analysts must be able to rely on long-lived isotopes if they are to gain any information from the sample.

ORIGEN-ARP Analysis

This project incorporated an comprehensive group of isotopes that are produced in a reactor, and the goal was to narrow that list to find those isotopic pairs best suited to determine history about the reactor cycle that produced the fission products. The ORIGEN-ARP simulations were run and analyzed every isotope produced in the reactor with a half-life greater than 60 days and a natural abundance less than 2%. As predicted, uranium and plutonium isotopes produced the highest R_c values. It was assumed that analysis of Pu and U would be most informative of the reactor cycle. However, this

project focuses on the case of reprocessed fuel in which the U and Pu have been removed.

An R_c value of 10^2 is taken to be sufficient, but many isotopic pairs are calculated to be well above this. The curium isotopic pairs appear to be more useful than plutonium, but curium is produced in such low quantities that its use for this work is not practical. Due to the assumption of a one year decay period, many of the isotopes useful in differentiating between cycles after a period of decay are those that are long-lived and thus best detected by AMS. Isotopes from the elements samarium, curium, and americium all have very promising R_c values, and may also be found in the detection calculation tables as producing an insufficient amount of counts for detection by decay spectroscopy.

Noticing a strong correlation between the long half-lives of isotopes in isotopic pairs and their consequent large cycle ratios, extensive simulations of the PWR, BWR, LMFBR, and the CANDU reactors were run with ORIGEN-2.2 concentrating on the isotopes detectable primarily by AMS. Simulations of both short and long cycles for each reactor design.

Values of R_c , R_d , and M_s were calculated for each of the isotopic pairs. When analyzing the data it was necessary to consider three separate cases:

- A) Initial knowledge of reactor design
- B) Initial knowledge of cycle length
- C) No initial knowledge of reactor design or cycle length

Consideration of each case produces different isotopic pairs that are most useful. It is necessary to consider these separate cases since the ratio values often intersect between differentiations of cycle length or reactor design; further explanation is found below.

Case A: Initial knowledge of reactor design

Evaluating a sample while knowing the reactor design in which the sample was created makes the cycle ratio values, R_c , extremely useful. Since we have simulated the behavior of 4 different reactors in this project, this case can be further separated into the 4 situations considering the PWR, BWR, CANDU, and LMFBR.

Table 2 lists the isotopic pairs that are most promising for AMS analysis to determine an approximate length of the reactor cycle in which they were created. The pairs differ depending upon reactor type. As previously stated the M_s value of the pair is taken as the larger of the two isotopes' M_s values. In many cases, a pair may have had a very large R_c but is not listed due to an extremely large M_s .

Table 2. (a) Cycle ratio and minimum sample size values for the PWR and BWR reactors corresponding to particular isotopic ratios. (b) Values for the CANDU and LMFBR reactors.

<u>PWR</u>			<u>BWR</u>		
<i>Isotopic Pair</i>	R_c	$M_s(\text{grams})$	<i>Isotopic Pair</i>	R_c	$M_s(\text{grams})$
$^{241}\text{Am}/^{243}\text{Am}$	9.22×10^2	6.05×10^{-5}	$^{241}\text{Am}/^{243}\text{Am}$	7.71×10^2	2.98×10^{-4}
$^{243}\text{Cm}/^{246}\text{Cm}$	3.08×10^3	7.21×10^1	$^{243}\text{Cm}/^{246}\text{Cm}$	1.24×10^3	1.05×10^3
$^{243}\text{Cm}/^{247}\text{Cm}$	1.00×10^5	9.02×10^4	$^{244}\text{Cm}/^{246}\text{Cm}$	5.03×10^2	1.05×10^3
$^{244}\text{Cm}/^{247}\text{Cm}$	2.14×10^4	9.02×10^4	$^{230}\text{Th}/^{229}\text{Th}$	4.05×10^2	7.90×10^2

$^{245}\text{Cm}/_{247}\text{Cm}$	1.68×10^3	9.02×10^4	$^{147}\text{Sm}/_{146}\text{Sm}$	3.03×10^2	3.05×10^{-1}
$^{244}\text{Cm}/_{246}\text{Cm}$	6.60×10^2	7.21×10^1	$^{149}\text{Sm}/_{146}\text{Sm}$	1.91×10^3	3.05×10^{-1}
$^{230}\text{Th}/_{229}\text{Th}$	1.00×10^3	4.03×10^2	$^{151}\text{Sm}/_{146}\text{Sm}$	7.14×10^2	3.05×10^{-1}
$^{147}\text{Sm}/_{146}\text{Sm}$	3.68×10^2	1.13×10^{-1}	$^{149}\text{Sm}/_{148}\text{Sm}$	2.18×10^2	1.06×10^{-7}
$^{149}\text{Sm}/_{146}\text{Sm}$	1.62×10^3	1.13×10^{-1}			
$^{151}\text{Sm}/_{146}\text{Sm}$	5.74×10^2	1.13×10^{-1}			
$^{149}\text{Sm}/_{148}\text{Sm}$	1.81×10^2	3.69×10^{-7}			

(a)

<u>CANDU</u>			<u>LMFBR</u>		
<i>Isotopic Pair</i>	R_c	$M_s(\text{grams})$	<i>Isotopic Pair</i>	R_c	$M_s(\text{grams})$
$^{241}\text{Am}/_{243}\text{Am}$	8.06×10^2	1.99×10^{-2}	$^{241}\text{Am}/_{242m}\text{Am}$	2.09×10^2	7.77×10^{-6}
$^{230}\text{Th}/_{229}\text{Th}$	1.11×10^2	7.22×10^3	$^{243}\text{Cm}/_{247}\text{Cm}$	8.73×10^2	7.20×10^2
$^{147}\text{Sm}/_{146}\text{Sm}$	3.04×10^2	1.12×10^1	$^{244}\text{Cm}/_{247}\text{Cm}$	1.06×10^4	7.20×10^2
$^{149}\text{Sm}/_{146}\text{Sm}$	2.94×10^3	1.12×10^1	$^{245}\text{Cm}/_{247}\text{Cm}$	5.29×10^2	7.20×10^2
$^{150}\text{Sm}/_{146}\text{Sm}$	1.66×10^2	1.12×10^1	$^{244}\text{Cm}/_{246}\text{Cm}$	4.76×10^2	3.52×10^{-1}
$^{151}\text{Sm}/_{146}\text{Sm}$	1.64×10^3	1.12×10^1	$^{230}\text{Th}/_{229}\text{Th}$	1.22×10^3	6.18×10^2
$^{149}\text{Sm}/_{148}\text{Sm}$	3.62×10^2	2.05×10^{-6}	$^{147}\text{Sm}/_{146}\text{Sm}$	2.53×10^2	5.82×10^{-2}
$^{151}\text{Sm}/_{148}\text{Sm}$	2.03×10^2	2.05×10^{-6}	$^{149}\text{Sm}/_{146}\text{Sm}$	3.38×10^2	5.82×10^{-2}
			$^{151}\text{Sm}/_{146}\text{Sm}$	3.77×10^2	5.82×10^{-2}

(b)

The pairs contained in each list are similar but sometimes vary significantly with respect to M_s or R_c . An analysis of each isotopic pair is not practical, but rather the list may serve as a guide to analysis depending upon the specific initial information that is known. For instance, a limit on sample size may be enforced therefore eliminating particular isotopic pairs, most likely those involving ^{246}Cm or ^{247}Cm . The possibility of analyzing a small sample size to ascertain specific historical information of the cycle is very real and is even more viable with the use of AMS.

Case B: Initial Knowledge of Cycle Length

The value of R_d is quite useful when prior knowledge of the sample's irradiation cycle is known. As with R_c , we were looking for orders of magnitude difference between the numerator and the denominator in R_d . This will allow for robust determinations to be made even if high sampling or detection errors exist.

R_d values for the ratio of PWR isotopes to BWR isotopes are all nearly equal to 1. This is expected due to the similarities of the PWR and BWR reactor cycle. The largest R_d values appear in isotopic pairs involving Uranium isotopes or Plutonium isotopes which are not our primary interest. In analysis of isotopes other than those of Uranium or Plutonium the utility of procedures to distinguish between reactor designs becomes scarce. Other than Uranium and Plutonium isotopes, there are no other isotopic pairs optimal for AMS that may definitively pinpoint one specific reactor for a measured ratio value.

Table 3. Isotopic pairs possessing $R_d > 10$. The similarities between the PWR and BWR severely limit the utility of R_d .

<i>Isotopic Pair</i>	<i>Reactor designs</i>	R_d	M_s (grams)
$^{150}\text{Sm}/^{146}\text{Sm}$	<i>PWR/CANDU</i>	3.38×10^{-02}	1.12×10^1
$^{150}\text{Sm}/^{146}\text{Sm}$	<i>PWR/LMFBR</i>	9.23×10^1	1.13×10^{-01}
$^{150}\text{Sm}/^{146}\text{Sm}$	<i>BWR/CANDU</i>	5.71×10^{-02}	1.12×10^1
$^{150}\text{Sm}/^{146}\text{Sm}$	<i>BWR/LMFBR</i>	1.56×10^2	3.05×10^{-01}
$^{150}\text{Sm}/^{146}\text{Sm}$	<i>CANDU/LMFBR</i>	2.73×10^3	1.12×10^1

Table 3 is an example of the primary problem that exists in case B, the close relation of the PWR and BWR. Although there may be some reasonable R_d values, the ratios of the PWR/ CANDU to the BWR/ CANDU are too close to distinguish between the BWR and PWR. It may be possible to determine the reactor type if the ratio is measured to correspond to the CANDU or LMFBR. Another reasonable scenario is that the analysts will not need to distinguish between the PWR and BWR due to their similar functionality; in this case many more isotopic pairs become useful when looking at the value of R_d .

Case C: No initial knowledge of cycle length or reactor design

This is the most important case since no assumptions are made about the amount of information that will be available to the analysts. Ultimately it is desirable for just one isotopic pair to tell us the whole story, *i.e.* to tell us from what type of reactor the sample came from, and how long that reactor cycle was. Unfortunately this is quite difficult and often the intersections of particular ratio values prevent this type of situation. Figure 1

illustrates the timelines of the isotopic pairs. Note that the values of R_c and R_d at any moment may easily be determined by comparing two different lifelines.

Figure 1 shows the graphical lifelines of each isotopic pair that meets the following parameters:

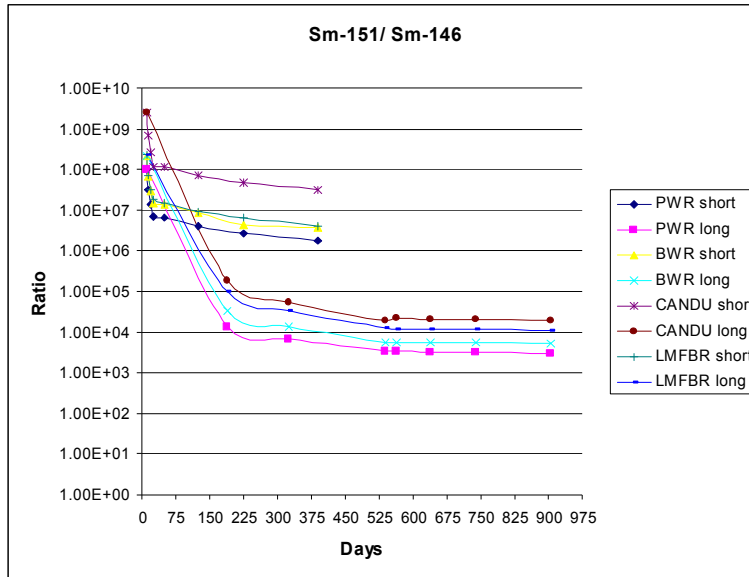
1. at least one of the isotopes is optimal for AMS
2. R_c of the pair $\geq 10^2$.
3. M_s of the pair $\leq 10^4$ grams *.

If a larger sample size may be obtained, for example an entire barrel of spent fuel, then more isotopes may be included and may form more valuable isotopic pairs. Each of the graphical interpretations of each isotopic pair were analyzed, but it is not plausible to definitively select a hierarchy of isotopic pairs for every situation. Such a hierarchy is highly dependent upon sample size obtained and the uncertainty involved in detection. In some instances, the ability to determine natural abundance of certain isotopes will be necessary so that isotopic pairs with stable isotopes, such as $^{146}\text{Sm}/^{149}\text{Sm}$, may be made.

Figure 1 shows the best six isotopic pairs for analysis in the situation of no initial knowledge and according to the above parameters under which the pairs were chosen. There is abundant graphical data for other isotopic pairs, however it would be impractical to discuss every pair. Figure 1a depicts the lifelines of the $^{151}\text{Sm}/^{146}\text{Sm}$ ratio for each reactor cycle. The short cycle is irradiated for 25 days and decayed for 1 year; the long cycle is irradiated for 540 days and decayed for 1 year. The close grouping between the lifelines of the long cycle and those of the short cycle show that the value of R_d between these reactors is not large. The ratio of the long and short ratio values can be easily

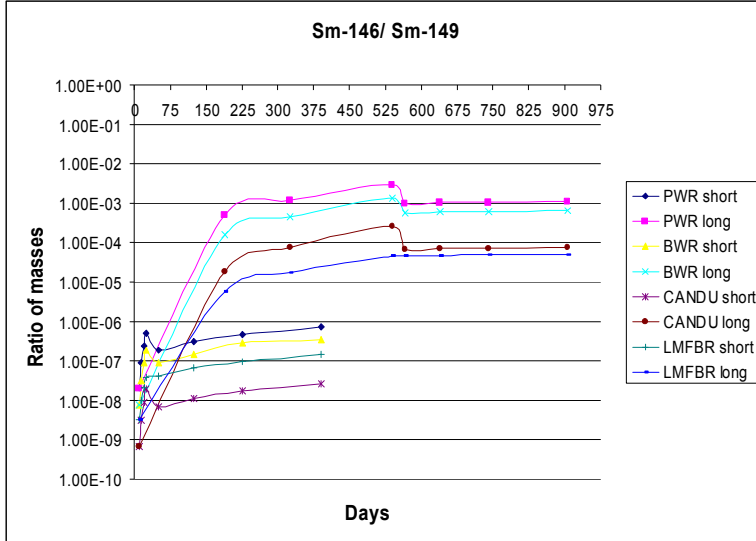
* ^{247}Cm and ^{246}Cm are included despite their $M_s \geq 10^4$ for the CANDU due the promising R_c values in the PWR, BWR, and LMFBR. However, the reader may note that insufficient amount of sample for detection can also be indicative of the CANDU design.

interpreted with the log scale of the y-axis. The 2 to 2.5 order of magnitude separation between the long and short cycle make this isotopic pair optimal for differentiating between cycle lengths. Another important factor of figure 1a is that conclusions are easily drawn from this pair's ratio values since none of the ratio values are shared amongst the short cycles and the long cycles.



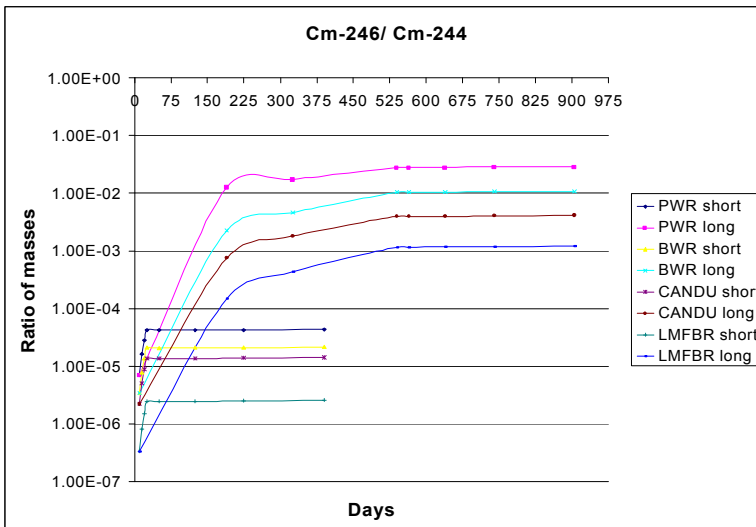
	PWR	BWR	CANDU	LMFBR
$\approx R_c$	10^2	10^2	10^3	10^2
$\approx M_s$	10^{-01}	10^{-01}	10^1	10^{-02}

(a)



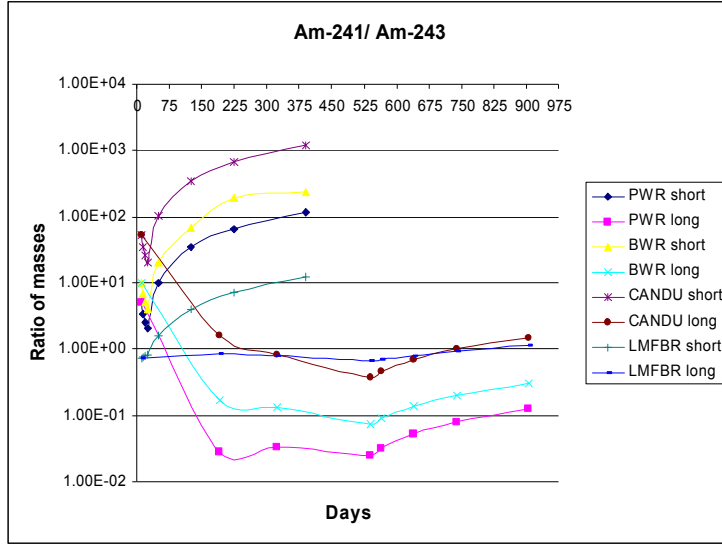
	PWR	BWR	CANDU	LMFBR
$\approx R_c$	10^3	10^3	10^3	10^2
$\approx M_s$	10^{-01}	10^{-01}	10^1	10^{-02}

(b)



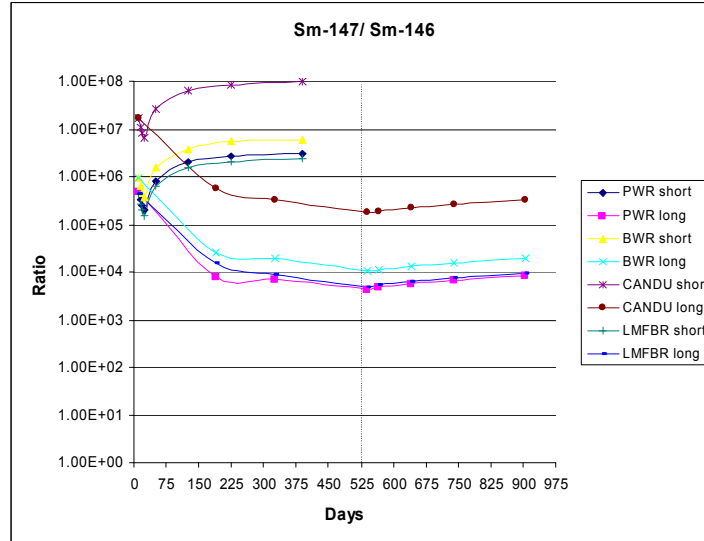
	PWR	BWR	CANDU	LMFBR
$\approx R_c$	10^2	10^2	10^2	10^2
$\approx M_s$	10^1	10^3	10^5	10^{-01}

(c)



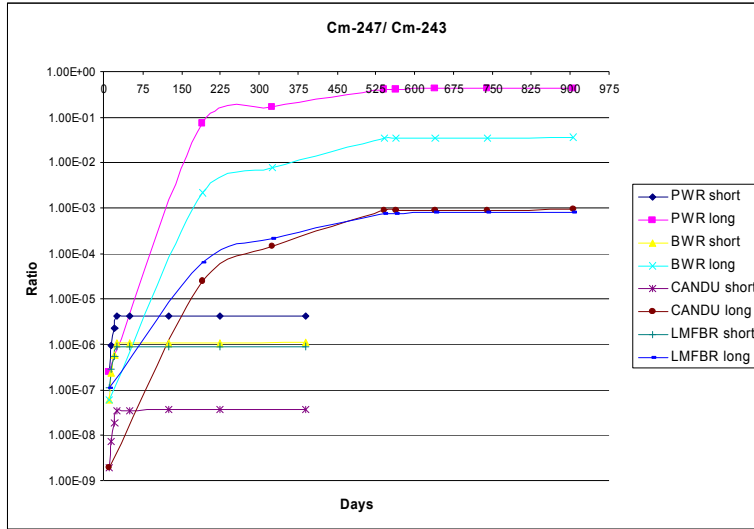
	PWR	BWR	CANDU	LMFBR
$\approx R_c$	10^2	10^2	10^2	10^1
$\approx M_s$	10^{-05}	10^{-04}	10^{-02}	10^{-09}

(d)



	PWR	BWR	CANDU	LMFBR
$\approx R_c$	10^2	10^2	10^2	10^2
$\approx M_s$	10^{-01}	10^{-01}	10^1	10^{-02}

(e)



	PWR	BWR	CANDU	LMFBR
$\approx R_c$	10^5	10^4	10^4	10^2
$\approx M_s$	10^4	10^6	10^9	10^2

(f)

Fig. 1. Graphical representation of isotopic pairs possessing large R_c values. Tables of R_c and M_s for each reactor design appear next the graph and time = 0 is the start of irradiation in the fuel cycle. (a) Sm-151/Sm-146 is very useful for cycle length differentiation. (b) ^{144}Sm measurement of natural abundance may also be necessary in order to determine amount of ^{149}Sm that is naturally in the sample when using this pair. (c) May encounter some analysis problems due to sample size with this pair, however an insufficient amount of sample for detection is also informative in some cases. (d) If detection uncertainty is low, this isotopic pair may be the most optimal. Detailed analysis of one sample may be informative of reactor design and cycle length. (e) Detection uncertainty is the limiting factor for analysis of this pair. (f) If a large sample size can be obtained, it is possible to determine cycle length and reactor design with analysis of this isotopic pair. However, the sample size limits any applicability towards the BWR and CANDU for this isotopic pair.

Conclusions

This work shows that analyzing a sample of spent fuel from a reactor for the purpose of deciphering historical characteristics of the reactor cycle is viable and is increased with the use of Accelerator Mass Spectroscopy. By gaining knowledge of the reactor cycle from which the isotopes originated one may gain insight into the purpose of the reactor's operation, *i.e.* for power or for other means. The cycle ratio values, R_c , reactor design ratio values, R_d , and sample size values, M_s , were developed to reduce the initially large list of isotopes down to the best suited isotopic pairs for fuel cycle analysis.

Three cases were discussed that were dependent on the amount of initial information known about the reactor cycle. R_c values of the isotopic pairs, $^{241}\text{Am}/^{243}\text{Am}$, $^{151}\text{Sm}/^{146}\text{Sm}$, and $^{243}\text{Cm}/^{246}\text{Cm}$ were strong indicators that these pairs would be very useful for fuel analysis to determine cycle length if the reactor design is known. Case B, which assumed initial knowledge of cycle length, was essentially based on the values of R_d for the isotopic pairs. The data showed that fuel analysis of isotopic pairs was not generally informative unless the analyst did not need to differentiate between the PWR and BWR.

Many suitable isotopic pairs were found to be useful for fuel analysis when no initial knowledge is present; the most suitable were found to be $^{151}\text{Sm}/^{146}\text{Sm}$, $^{149}\text{Sm}/^{146}\text{Sm}$, and $^{244}\text{Cm}/^{246}\text{Cm}$. However, it is necessary for the analyst to choose the most suitable isotopic pairs depending on specific initial conditions, and possibly even more importantly, the available sample size.

The isotopic pairs chosen to be most suitable from this project's research are based upon the assumption that AMS is an available resource to the analyst. In many

circumstances, one or both of the isotopes in a pair will not be detectable by traditional spectroscopy due to their long half-lives or low gamma yield. Such characteristics are prominent in surviving isotopes of spent fuel that has been decaying for an unknown amount of time. Consequently, the ability to perform mass spectrometry is increasingly beneficial when one's ignorance of the initial conditions of the sample is significant.

Table 4 lists the isotopes that are considered to be the most useful for AMS analysis to determine historical characteristics of a nuclear fuel cycle from which the isotope originated. The isotopes are grouped together according to their atomic numbers. Any box that is marked specifies that some pair of those isotopes can be useful for determining information denoted by the box.

Acknowledgement

This work was performed in part under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

Isotope	Cycle Length Determination								Reactor Design Determination							
	Know Reactor Design				No Initial Knowledge				Know Cycle Length				No Initial Knowledge			
	PWR	BWR	CANDU	LMFBR	PWR	BWR	CANDU	LMFBR	PWR	BWR	CANDU	LMFBR	PWR	BWR	CANDU	LMFBR
²⁴¹ Am, ^{242m} Am, ²⁴³ Am	X	X	X	X												
²⁴³ Cm, ²⁴⁴ Cm, ²⁴⁵ Cm, ²⁴⁶ Cm, ²⁴⁷ Cm, ²⁴⁸ Cm	X	X	X	X	X	X	X	X								
¹⁴⁶ Sm, ¹⁴⁷ Sm, ¹⁴⁸ Sm, ¹⁴⁹ Sm, ¹⁵⁰ Sm, ¹⁵¹ Sm	X	X	X	X	X	X	X	X							X	
²²⁹ Th, ²³⁰ Th	X	X	X	X	X	X	X									

Table 4. The above table is a broad summary of the applicable uses that certain isotopes of Americium, Curium, Samarium, and Thorium may be useful for as indicators of reactor fuel cycle characteristics. It was determined that it is extremely difficult to differentiate between the four reactor designs due to the similarities of the PWR and BWR.

References

1. X.D. HOU, W. CHEN, Y.H. HE, and B.T. JONES, "Analytical atomic spectrometry for nuclear forensics." *Appl. Spectros. Rev.*, **40**, 245 (2005).
2. J.S. VOGEL, K.W. TURTELTaub, R. FINKEL, and D.E. NELSON, "Accelerator Mass-Spectrometry - Isotope Quantification At Attomole Sensitivity," *Anal. Chem.*, **67**, A353 (1995).
3. W. KUTSCHERA, "Progress in isotope analysis at ultra-trace level by AMS," *Int. J. Mass Spectrom.*, **242**, 145 (2005).
4. D. MCCURDY, Z. LIN, K.G.W. INN, R. BELL, S.WAGNER, D.W. EFURD, R. STEINER, C. DUFFY, T.F. HAMILTON, T.A. BROWN, and A.A. MARCHETTI, "Second interlaboratory comparison study for the analysis of Pu-239 in synthetic urine at the mu Bq (similar to 100 aCi) level by mass spectrometry," *J. Radioan. Nucl. Chem.*, **263**, 447 (2005).
5. T. STRAUME, G. RUGEL, A.A. MARCHETTI, W. RUHM, G. KORSCHINEK, J.E. MCANINCH, K. CARROLL, S. EGBERT, T. FAESTERMANN, K. KNIE, R. MARTINELLI, A. WALLNER, and C. WALLNER, "Measuring fast neutrons in Hiroshima at distances relevant to atomic-bomb survivors," *Nature* **424**, 539 (2003).
6. M. PAUL, D. BERKOVITS, L.D. CECIL, H. FELDSTEIN, A. HERSHKOWITZ, Y. KASHIV, and S. VOGT, "Environmental Sr-90 measurements," *Nucl. Instr. & Meth. B* **123**, 394 (1997).
7. L. WACKER, L.K. FIFIELD, and S.G. TIMS, "Developments in AMS of Tc-99," *Nucl. Instr. & Meth. B*, **223**, 185 (2004).
8. R. MICHEL, J. HANDL, T. ERNST, W. BOTSCH, S. SZIDAT, A. SCHMIDT, D. JAKOB, D. BELTZ, L.D. ROMANTSCHUK, H.A. SYNAL, C. SCHNABEL, and J.M. LOPEZ-GUTIERREZ, "Iodine-129 in soils from Northern Ukraine and the retrospective dosimetry of the iodine-131 exposure after the Chernobyl accident," *Sci. Total Environ.* **340**, 35 (2005).
9. U.P. JENQUIN, R.J. GUENTHER, "Evaluation of ORIGEN2 models against spent-fuel radiochemical measurements," *Transactions of the American Nuclear Society*. **61**, 76 (1990).
10. T. ADACHI, Y. NAKAHARA, N. KOHNO, K. GUNJI, T. SUZUKI, T. SONOBE, M. ONUKI, K. KATO, E. TACHIKAWA, S. INOUE, M. TAKAYASU, M. YOSHIKUNI, S. KOBAYASHI, Z. TERUYA, T. KAWAMOTO, and M. KAWAMURA, "Comparison of calculated values with

- measured values on the amount of TRU and FP nuclides accumulated in gadolinium bearing PWR spent fuels,” *J. Nucl. Sci. & Tech.*, **31**, 1119 (1994).
11. W.S. CHARLTON, B.L. FEARE, C.W. NAKHLEH, T.A. PARISH, R.T. PERRY, J. POTHS, J.R. QUAGLIANO, W.D. STANBRO, and W.B. WILSON, “Operator Declaration Verification Techniques for Spent Fuel at Reprocessing Facilities,” *Nucl. Instr. & Meth. B*, **168**, 98 (2000).
 12. O.W. HERMANN, M.D. DEHART, “Validation of SCALE (SAS2H) Isotopic Predictions for BWR Spent Fuel,” *Oak Ridge National Lab/TM-13315*, (1998).
 13. I.C. GAULD, K.A. LITWIN, “Verification and Validation of the ORIGEN-S Code and Nuclear Data Libraries,” *Atomic Energy of Canada Limited/RC-1429*, (1995).