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Trace Fission Product Ratios for Nuclear Forensics Attribution of Weapons-Grade Plutonium from Fast and Thermal Reactors

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The growing concern about nuclear terrorism threats has enhanced the need to develop fast and accurate nuclear forensics analysis techniques for nuclear material source attribution and to create a credible nuclear deterrence. Plutonium produced as a by-product in nuclear reactor fuel, especially in fuel discharged at low burn-up (1 to 2 MWd/kg), is potentially weapons usable material. In the event of plutonium interdiction from a smuggling act, its origin has to be established through nuclear forensics attribution methods before any response is initiated against this malicious act. The characteristics of separated plutonium from discharged reactor fuel and the associated fission product traces depend on factors such as the reactor type (thermal or fast reactor), fuel burn-up, irradiation history, and the chemical process used to separate plutonium. A new methodology of using trace fission product to plutonium ratios for nuclear forensics attribution of plutonium to the type of reactor used for its production is presented along with results obtained for case studies of a fast neutron spectrum breeder reactor and a thermal neutron spectrum reactor using open literature design information of these two types of nuclear reactors.

INTRODUCTION

The Nuclear Forensics and Attribution Act, signed into law by U.S. President Barack Obama in 2010, states that a nuclear terrorist attack is one of the most serious threats to the national security of the United States.[1] Nuclear

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safeguards methods are used to verify the use of nuclear materials for peaceful purposes, and to deter the proliferation of nuclear weapons by detection of the diversion of special nuclear materials and misuse of technologies. The major risk of nuclear weapons' proliferation however, lies with countries that have significant unsafeguarded nuclear activities.[2] The threat of nuclear materials and possible weapons produced from unsafeguarded activities illustrates the value of a robust nuclear forensics capability, enabling nuclear material attribution to its origin.

Plutonium, a byproduct in spent nuclear fuel, is generated in nuclear reactors from uranium through a nuclear reaction. The production of plutonium-239 is through the neutron capture reaction of uranium-238 followed by two beta decays.

$$^{238}_{92}\mathrm{U} + ^{1}_{0}\mathrm{n} \xrightarrow{(\mathrm{n},\gamma)} ^{239}_{92}\mathrm{U} \xrightarrow{\beta^-} ^{239}_{93}\mathrm{Np} \xrightarrow{\beta^-} ^{239}_{94}\mathrm{Pu} \tag{1}$$

Fuel burn-up is defined as the thermal energy produced per unit mass of nuclear fuel. As fuel burn-up increases, neutron capture reactions in plutonium-239 and successive isotopes of plutonium lead to the buildup of higher mass number plutonium isotopes. Eventually a full composition of plutonium isotopes (plutonium-238, plutonium-239, plutonium-240, plutonium-241, and plutonium-242) exists in the irradiated fuel. A higher concentration of plutonium-239 translates to higher quality plutonium for use in nuclear weapons. For burn-up levels commonly achieved in nuclear power reactors, the resulting plutonium is reactor-grade (approximately 60 percent plutonium-239). It is known though, that weapons-grade plutonium (approximately 94 percent plutonium-239) will be produced if uranium is subject to a low burn-up of about one to two megawatt days per kilogram (MWd/kg).[3]

Normally, the International Atomic Energy Agency (IAEA), through the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) and safeguards agreements with countries, monitors such plutonium. However there are cases of plutonium production occurring in states where nuclear fuel cycle facilities are not under IAEA safeguards.[4] In this aticle the authors study the characteristics of plutonium and the associated trace element contaminants when separated from fuel discharged at a very low burn-up from fast neutron spectrum breeder and thermal neutron spectrum reactors. Both of these reactor types have the ability to produce weapons-grade plutonium. Because the core design data needed for this study is readily available in the open literature, India's 500 megawatt electric (MWe) prototype fast breeder reactor (PFBR) is taken as an example for the fast neutron spectrum reactor core,[5] and a pressurized heavy water reactor (PHWR) is used as an example for the thermal neutron spectrum reactor core.[6]

Fast breeder reactors are designed to produce plutonium in the blanket material (uranium-238). Such reactors, however, can be operated to generate

low burn-up, weapons-grade plutonium. The PFBR, in the advanced stage of construction, will produce significant quantities of weapons-grade plutonium during operation. Previous work has estimated that about 140 kilogram (kg) of weapons-grade plutonium will be produced in the blankets of the PFBR each year.[7]

The PHWRs are of particular interest because of their unique online refueling capabilities. Light water reactors are loaded with large excess reactivity initially and therefore need to undergo batch refueling once every 12 to 18 months. The batch refueling requires the reactor to be shutdown, which makes frequent refueling from a commercial or operational point of view less attractive. In contrast, heavy water reactors are frequently refueled while online and in operation. The low reactivity of natural uranium fuel leads to typically refueling one fuel channel per day. The PHWRs usually reach an average fuel burn-up of about 6.7 MWd/kg.[8] However, online refueling makes the reactors more susceptible to the diversion of material from the core. This introduces the potential for the fuel to be intentionally exposed to a low burn-up and then removed from the core for use outside of civilian energy production.

There are several differences between the PFBR and PHWR including neutron spectrum, fuel material, moderator and coolant, all of which may lead to variances in the burned fuel composition. Thus, the resulting composition of the discharged fuel may be able to provide information on the reactor system which produced the plutonium.

The objective of this study is to estimate the concentrations of plutonium and fission product isotopes in spent fuel from the PFBR peripheral radial blanket sub-assemblies and PHWR low burnt fuel bundles, and to develop a suite of isotopic ratios useful for quick and accurate nuclear forensics attribution of the source reactor for interdicted weapons-grade plutonium. The plutonium and fission product estimations were obtained through developing three-dimensional reactor core models and by performing neutron transport simulations and fuel burn-up calculations using Monte Carlo radiation transport methodology. The following sections discuss previous studies in this subject area, details of the current study, results, and their relevance to nuclear forensics methods.

PREVIOUS NUCLEAR FORENSICS STUDIES

Multiple studies on technical nuclear forensics have been published which demonstrate the ability of isotopic data to retain information about the source of the produced special nuclear material. An environmental monitoring system was developed at Los Alamos National Laboratory (LANL) using fissiogenic noble gases, namely xenon and krypton, as a verification technique for reprocessing facilities.[9] The relative concentrations of stable xenon and krypton isotopes depend on several reactor operating parameters. Measurements

of isotopic ratios of these noble fission gases could thus be used to verify operator declarations or determine fuel parameters such as fuel type, burn-up, and reactor type. The calculated database of xenon and krypton isotopic ratios was created using a series of reactor analysis codes to model essentially all types of reactors. Computational modeling showed the ability of isotopic ratios to distinguish between light water reactors, heavy-water reactors, and breeder reactors. This system utilizes the fact that noble gases are not chemically bound to the fuel and are thus released during reprocessing. Although the isotopic ratios and database developed by this technique are not useful for analyzing post-processed materials, it demonstrates that fission product concentrations carry the information needed to infer a source reactor.

A master's thesis by M. R. Scott developed a forensics methodology for attributing spent fuel used in a radiological dispersal device to a source reactor.[10] The attributes included the spent fuel burn-up, age after discharge, reactor type, and initial fuel enrichment. To find a distinction between reactor types, isotopes with cross sections and yields that change significantly between reactor types were needed. Ratios of the chosen isotopes were plotted against burn-up for each reactor. The results showed that these isotopes could easily differentiate between a fast reactor and thermal reactor. Scott's work, however, was focused on higher burn-up levels that are more commonly achieved in power reactors, rather than low burn-up weapons-grade plutonium. Isotopes coming from more complex production chains do not have sufficient time to be produced in low-burn-up material. As a result, the monitors identified in Scott's study were not suitable for the nuclear forensics analysis of low-burn-up fuel, the subject presented here.

A previous study was completed by Wallenius et al. in which plutonium isotopics were analyzed and used for the purposes of origin determination of plutonium seized in the illicit trafficking of nuclear material.[11] This study used the reactor fuel burn-up code, ORIGEN2,[12] to calculate the plutonium composition for multiple reactors, as well as a thermal ionization mass spectrometer to measure plutonium isotope ratios of five plutonium samples. The sources of the plutonium samples included two from the National Bureau of Standards, two from the former Soviet Union, and one sample from an International Technical Working Group (ITWG) round robin test. Following measurements, a source reactor for each sample was inferred by comparing the measured and computationally calculated isotopic compositions. Their study raises a number of concerns with regards to the isotopics and computer modeling. The isotopic analyses consist of plutonium and actinides only, with no investigation of contaminant fission products. The isotope generation and depletion code, ORIGEN2, uses a zero-dimensional fuel model giving the composition averaged over the whole reactor core. This could be problematic with a fast breeder reactor (FBR), whose core consists of very different fuel and blanket regions. Additionally, irradiation times were considered to be

continuous and no cooling time corrections were made to the material. Burn-up levels obtained for the computational models were equivalent to typical burn-up for each reactor with the exception of the FBR. Here, the FBR blanket material has a burn-up of 20 MWd/kg, which is relatively high. This burn-up level is likely due to the averaging of core and blanket fuel, as a result of ORIGEN2 modeling.

Glaser studied the isotopic signatures of weapons-grade plutonium produced in reactor types which have been historically used for plutonium production.[13] Three types of reactors, the Hanford-type, NRX-type, and Calder Hall-type reactors were modeled. For each of these reactor types, the plutonium composition was obtained and ratios of plutonium isotopes were analyzed. It was determined that predictive signatures derived from the plutonium isotopic ratios can distinguish weapons-grade plutonium from basic reactor types including FBRs, light water reactors using low-enriched fuel, and reactors fueled with natural uranium. Fission products are absent in the analysis done. While a forensics methodology consisting purely of plutonium isotope ratios could be beneficial due to independence from the separation technique, the plutonium isotopics may be assisted by the inclusion of fission product compositions.

Previously published studies have developed plutonium or fission product isotope analysis techniques which attribute nuclear material to a source reactor. Most of the research completed however, has been focused on reactor spent fuel, irradiated typically to an average burn-up level, where the composition of plutonium is not weapons-grade. Lacking are investigations into isotopic characteristics of fission product contaminants in separated weapons-grade plutonium as a result of low burn-up fuel from reactor misuse or a breeder reactor blanket. Using ratios of fission products to plutonium in separated weapons-grade plutonium for nuclear forensics isotope analysis, as presented here, is a novel approach for source reactor attribution.

METHODOLOGY

A detailed characterization of weapons-grade plutonium includes fission product contaminants and plutonium isotopics. The potential for trace amounts of fission products is due to the non-ideal chemical process used to separate plutonium from irradiated nuclear fuel. The degree of purification achieved by a separation process can be quantified by a decontamination factor (DF), which is the ratio of a stated impurity to the desired component in the feed divided by the equivalent ratio in the product.[14]

$$DF = \frac{\left[\frac{Impurity}{Desired\ Component}\right]_{Feed}}{\left[\frac{Impurity}{Desired\ Component}\right]_{Product}} \tag{2}$$

The most commonly employed technique for plutonium separation is the plutonium uranium recovery by extraction (PUREX) process.[15] Using the PUREX process, decontamination factors of 10^6–10^8 have been achieved for the reduction of fission products in separated plutonium, however a measurable contaminant will remain.[16]

Similar to the plutonium composition, fission product inventories are dependent on parameters such as fuel burn-up, the type of fuel, the enrichment, the neutron energy spectrum of the reactor, and the time after irradiation. The objective of this research is to analyze plutonium and fission products from the PFBR radial blanket fuel with a burn-up of 1 MWd/kg, as well as PHWR bundle fuel with low burn-up of 1 and 2 MWd/kg. For these reactor fuels exposed to equal levels of burn-up, the drastically different neutron energy spectra will be the source of variations in fission product inventories. The dissimilarity of the neutron spectra lead to different amounts of plutonium and fission product concentrations in the discharged fuel even though the fuel burn-up is equal. Discrepancies result from variations in the fission yield for several fission products, variations in neutron interaction cross-sections for fission product isotopes, and variations in interaction cross-sections for plutonium isotopes.

Prototype Fast Breeder Reactor

Detailed information for the 500-MWe Indian PFBR, including essential design parameters and operation procedures were obtained from the literature.[17] An active core, one meter in height, consists of an inner and outer core of mixed-oxide (MOX) driver fuel. The MOX fuel has plutonium oxide content of 20.7 percent for the inner core and 27.7 percent for the outer core. This increases the amount of fissile material around the periphery of the active core, thus flattening the neutron flux profile across the reactor. Axial blankets of length 0.3 meters (m) each, sit above and below the active core, all of which are surrounded by 1.6 m tall radial blankets. Both core blankets are comprised of depleted uranium oxide target fuel, capturing the neutrons leaking from the core. This large amount of uranium-238 is where the plutonium breeding will take place. The plutonium bred in the axial and radial blankets are likely to have similar characteristics, however this project is focused on the plutonium produced in the radial blankets only. A core map of the PFBR, in Figure 1, displays the inner core, outer core, radial blanket sub-assemblies, control safety rods (CSR), and diverse safety rods (DSR), in the equilibrium core configuration. Figure 2 gives a cross-sectional view of inner and outer core and radial blanket sub-assemblies.

The standard operating scheme of the PFBR is an 180 day cycle of full power operation followed by 60 days for shutdown and refueling, with one third of the active core being refueled at the end of each cycle. Conversely, the radial blanket sub-assemblies are refueled with a slightly different pattern, but still

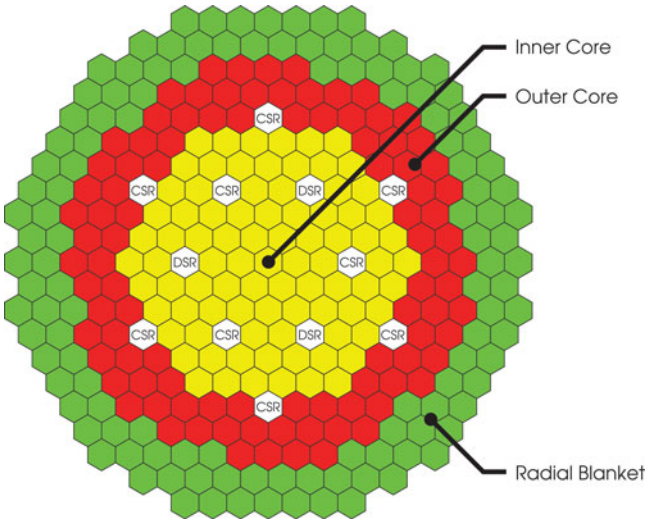

Figure 1: Core map of the 500-MWe Indian prototype fast breeder reactor.

on the basis of fuel burn-up. Typical operation of the PFBR will be to discharge the radial blanket sub-assemblies during the refueling stage in which the burn-up is nearest to 1 MWd/kg. Thus the radial blanket is split into three sections. Forty-two blanket sub-assemblies, which are in close proximity to the core and are therefore exposed to a larger neutron flux, are replaced after every 180 day cycle. Six radial blanket sub-assemblies are refueled after every two cycles, and seventy-two blanket sub-assemblies that are located farther from the core are irradiated for three cycles before being refueled.

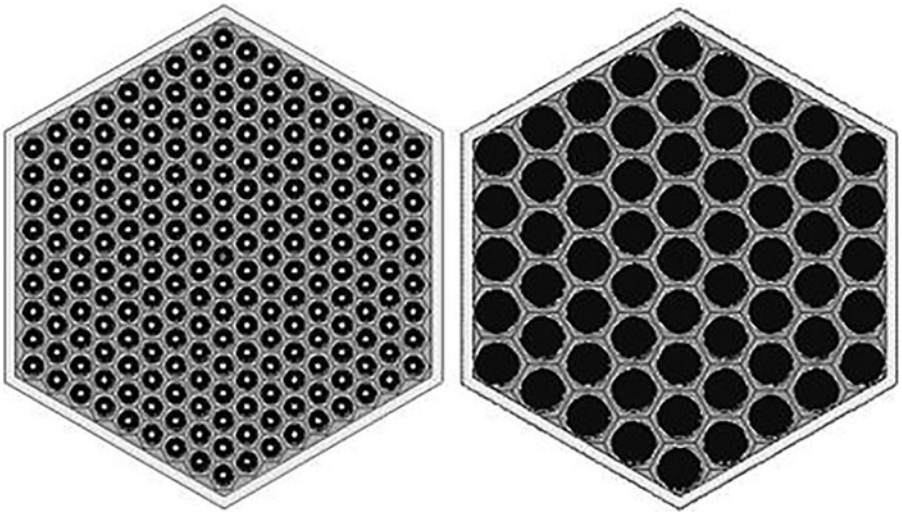

Figure 2: Fuel pin arrangement for core and radial blanket sub-assemblies of the prototype fast breeder reactor.

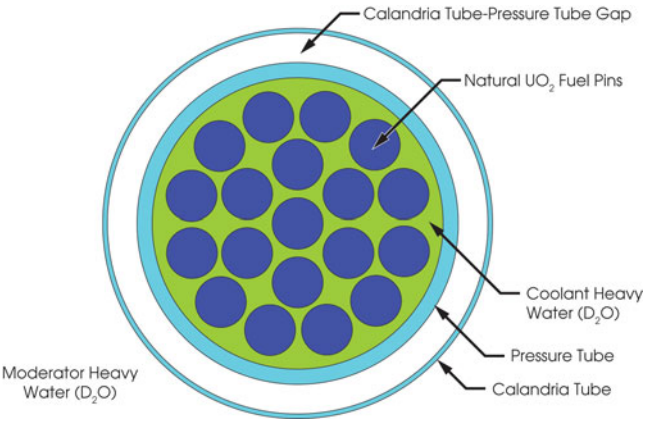

Figure 3: Cross-sectional view of Indian pressurized heavy water reactor 19-pin fuel bundle model.

Pressurized Heavy Water Reactor

Details of the Indian PHWR (220 MWe–756 MWth) used to develop the reactor-fuel bundles model for the fuel burn-up simulations were obtained from the literature.[18] The Indian PHWR is a heavy water (deuterium oxide) moderated, heavy water cooled horizontal pressure tube type power reactor with 306 coolant pressure tubes. Each pressure tube holds 12 fuel bundles each with a length of 490 millimeters (mm). Fuel bundles are comprised of 19 natural uranium dioxide fuel pins clad in Zircaloy and arranged in three rings as shown in Figure 3.

The PHWRs are refueled on-power because the fuel worth of uranium dioxide of natural isotopic composition (i.e., 0.7 percent of the uranium consist of the fissile isotope uranium-235) depletes faster than the uranium dioxide fuel used in light water reactors, which is enriched up to 3–4 percent in uranium-235. On an average, 10 fuel bundles are refueled per effective full power day (EFPD) of operation.[19] During the refueling operation, a pre-identified coolant pressure tube location is chosen based on core physics calculations. Four bundles in the pressure tube are reshuffled and remain in the channel once the refueling is complete, while the remaining eight bundles are removed and replaced with fresh fuel bundles.[20]

Modeling of Reactor Cores, Radiation Transport Calculations and Fuel Burn-up Simulations

Reactor core burn-up simulations were performed utilizing the Monte Carlo radiation transport code, MCNPX Version 2.7.[21] The burn-up/depletion code, CINDER90, comes built-in to the MCNPX-2.7 package.[22] A detailed pin-by-pin three dimensional model of the PFBR core was created to simulate the

reactor through cycles of 180 days of full power operation followed by 60 days of shutdown and refueling. Multiple successive inputs were built for the reactor, each having material comprised of the predecessor's output, to accurately simulate the refueling and core alterations as it reached an equilibrium core configuration. These computer simulations provided an estimate of the plutonium composition and fission product composition within the discharged PFBR blanket fuel.

The online refueling aspects of the PHWR described above were also incorporated into the simulations for the fuel bundle to reach low-burn-up levels of 1 MWd/kg and 2 MWd/kg. In addition to the whole core simulations, fuel burn-up simulations using a single fuel bundle, as seen in Figure 3, with reflective boundary conditions and a lattice pitch of 228.6 mm were also performed. The changes in the estimated fission product and plutonium isotope concentrations for isotopes of interest to this study were found to be less than 2 percent between the single bundle lattice level simulation and the whole core simulation. Hence, the less computationally intensive single bundle lattice level simulations were performed to estimate the plutonium and fission product concentrations in the discharged PHWR bundle fuel.

RESULTS AND DISCUSSIONS

Neutron Energy Spectrum

As mentioned above, the neutron spectrum to which the fuel is exposed has a large effect on the plutonium and fission product isotopic concentrations present in the spent material. The 44 energy group neutron spectra obtained from MCNPX-2.7 simulations for the inner core of the PFBR and the PHWR single bundle are plotted in Figure 4.

Weapons-grade Plutonium Production

The significant quantity (SQ) of plutonium, defined as the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded, is 8 kg.[23] Estimations for the amount of plutonium produced, as well as isotopic composition, in the low burn-up fuel of the two reactor types were obtained using MCNPX-2.7 burn-up simulations. Results from the simulations are shown in Tables 1 and 2.

The radial blanket is divided into three refueling groups, in order to remove the material when the burn-up is nearest 1 MWd/kg. Table 1 provides the results from each group of the radial blanket. The groups are titled by the number of cycles spent in the reactor when removed. The group of radial blanket assemblies which see three irradiation cycles come closest to the target

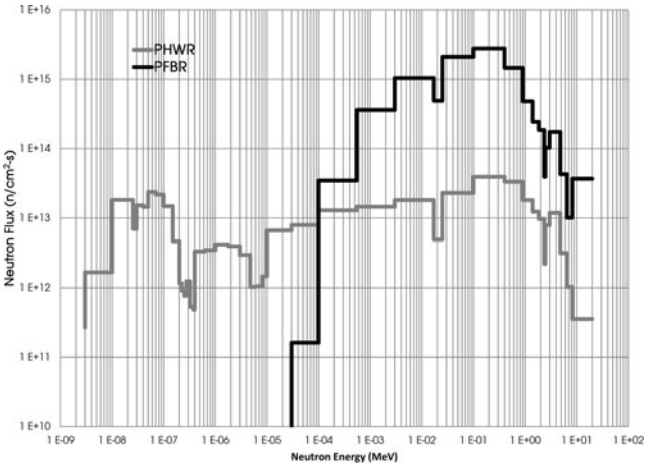

Figure 4: Neutron energy spectra from the prototype fast breeder reactor (PFBR) and pressurized heavy water reactor (PHWR).

burn-up of 1 MWd/kg. The results from this "three cycles" group of radial blanket assemblies are the results used for the selected isotopes described later.

Table 2 gives the plutonium composition calculated in the PHWR single fuel bundle at burn-ups of 1 MWd/kg and 2 MWd/kg. Results in Table 2 show that plutonium produced at both of the low fuel burn-up levels is weapons-grade. The plutonium-238 concentration was less than 0.01 percent for both burn-up levels. Although only about 10 to 20 grams of plutonium is produced in a single fuel bundle, it is worth noting that there are 3,672 fuel bundles in the PHWR core.

Development and Selection of Isotopic Ratios

An analysis of various isotopes is necessary to link material to a source reactor. The ultimate goal being the development of a suite of isotopic ratios capable of attributing the source of separated weapons-grade plutonium. Selected isotopes are reported in Tables 3 and 4, as the expected mass and radioactivity of each isotope which would be present in 1 kg of separated plutonium. One benefit to reporting the selected isotopes as ratios per mass of plutonium is the ability to scale the data to the mass of the interdicted weapons-grade plutonium. For experimental investigations, a few micrograms of plutonium are sufficient to detect the fission product contamination. Selection was based on (a) the amount of isotope production, at least a few picograms per kg of plutonium, (b) the probability of detection (high gamma energy greater than 100 keV, long half-life greater than 100 days, high radioactivity greater than 1 microcurie), (c) reactor type dependence in isotope production, and (d) the PUREX plutonium reprocessing decontamination factor (DF) of the isotope.

Table 1: Plutonium produced in prototype fast breeder reactor radial blanket regions.

Irradiation Time (180-days Cycles)	No. of Assemblies	Burn-up (MWd/kg)	Total Mass of Plutonium (kg)	Plutonium Isotopes (%)				
				Plutonium -238	Plutonium -239	Plutonium -240	Plutonium -241	Plutonium -242
3	72	1.017	99.15	<0.01	98.02	1.93	0.04	<0.01
2	6	1.3	8.67	<0.01	98.18	1.78	0.03	<0.01
1	42	0.71	34.87	<0.01	98.97	1.02	0.01	<0.01

Table 2: Plutonium produced in a single fuel bundle of the pressurized heavy water reactor.

Irradiation Time (Days)	Burn-up (MWd/kg)	Total mass of Pu (grams)	Plutonium Isotopes (%)				
			Plutonium -238	Plutonium -239	Plutonium -240	Plutonium -241	Plutonium -242
24 (and 60 days decay)	1.012	10.23	<0.01	95.98	3.78	0.23	0.01
48 (and 60 days decay)	2.025	21.96	<0.01	91.54	7.58	0.83	0.04

Specific decontamination factors on an elemental basis could not be found in open literature, hence a DF of 10^6 was applied universally. As the same DF is applied to all isotopes, with the exception of plutonium, it is still an important characteristic. Given that the material is separated, fission products will be reduced to trace contaminants in the nearly pure plutonium. For fission products with a small amount of production, the inclusion of a DF from separation, results in levels which are undetectable. In Tables 3 and 4, the isotopes are classified into four groups namely, prompt gamma, alpha, other gamma, and mass spectrometry, based on the detection method and how fast results can be obtained. Results can be acquired in a few hours with gamma spectroscopy, whereas it can require days or weeks for alpha spectroscopy and mass spectrometry processes, respectively.

Tables 3 and 4 contain the expected mass and activity of selected isotopes within a kilogram of separated weapons-grade plutonium produced from the PFBR and PHWR. The ratio of isotope mass per unit plutonium from the PHWR divided by the isotope mass per unit plutonium from the PFBR, both with a fuel burn-up of 1 MWd/kg, depicts the isotope ratio's reactor dependence. These values are given in Table 5. Several important observations from Table 3, 4, and 5 are discussed in the following paragraphs.

The radioactivity concentration of cesium-137 and cerium-144 isotopes are sufficiently high in 1 kg of plutonium and gamma spectroscopy measurements can be made quickly (prompt measurements) once such material has been interdicted. Both cesium-137 and cerium-144 undergo beta radiation decay followed by gamma emissions of 662 keV and 134 keV, respectively. The commonly used burn-up monitor, cesium-137, is an interesting isotope to note when used to display a reactor dependency.

It was found that a selected fission product ratio to plutonium provides more information and results in larger differences between reactors, than just the absolute abundance of a particular isotope. The radioisotope cesium-137, for example, is an attractive isotope for selection. The individual fission yield is high at around 6 percent, it has a long half-life of over 30 years, and the gamma radiation is easily measurable. However, cesium-137 is a direct fission product with a fission yield that is constant regardless of fissile isotope or neutron energy. The amount of cesium-137 can provide information on the burn-up of a material but no information regarding the source reactor. The ratio of cesium-137 to plutonium, though, is found in this study to result in a significant difference between the PFBR and PHWR. The ratios of fission products to plutonium have the ability to distinguish between fast and thermal reactors. This is due, in large part, to the amount of plutonium the PFBR breeds. The PFBR has a larger percentage of uranium-238 in the depleted uranium fuel in addition to the effect of an intense fast neutron spectrum. Thus the PFBR radial blanket produces much more plutonium per initial loading of uranium (approximately 1 percent) than the PHWR (approximately 0.1 percent).

The radioactivity of plutonium-239 and plutonium-242 isotopes is sufficiently high in 1 kg of plutonium and alpha spectroscopy measurements can be made. However, sample preparations are needed for performing alpha spectrometry, which makes this method slower than prompt gamma radiation measurements. Both plutonium-239 and plutonium-242 undergo alpha decay with energies of 5156 keV and 4901 keV, respectively. These alpha energies are distinct enough to be uniquely identified in the alpha spectra. A fast or thermal neutron spectrum can likely be determined from an alpha or mass spectrometry measurement of plutonium-239 and plutonium-242, alone. The PHWR to PFBR ratio of plutonium-239 concentration is 0.98, while the ratio of plutonium-242 concentration is 19.15. This indicates that much less plutonium-242 is present in the plutonium produced in the fast spectrum. This is a result of the relative differences in neutron interaction cross-sections between absorption and fission at varying neutron energies. Lower concentrations of heavier plutonium isotopes, specifically plutonium-241 and plutonium-242, are present in the PFBR blanket fuel due to fission being more likely than radiative capture at fast neutron energies.

The next set of isotopes, cesium-134, antimony-125, and europium-154 is again proposed to be measured via gamma spectroscopy. The radioactivity concentrations for these gamma emitting isotopes, however, are orders-of-magnitude less than the prompt gamma measurement isotopes, cesium-137 and cerium-144. Mass spectroscopy is anticipated as the measurement technique for rubidium-85, strontium-90, neodymium-148, promethium-147, and samarium-150. The isotopes rubidium-85, neodymium-148, and samarium-150 are stable and are thus undetectable using radiation measurements. The isotopes strontium-90 and promethium-147 are pure beta radiation emitters without any gamma energy emissions. Of the isotopes proposed to be measured using mass spectroscopy, samarium-150 is particularly significant. The PHWR to PFBR ratio of samarium-150 concentration is 107, meaning plutonium produced in a thermal neutron spectrum will have two orders-of-magnitude more samarium-150 contamination than plutonium produced in a fast spectrum. The source of this large difference is a result of the radiative capture cross-section of the well-known fission product neutron poison, samarium-149. The plot of the samarium-149 radiative capture cross-section per incident neutron energy is shown in Figure 5.[24] The dominant neutron energy of the PFBR is 100 keV to 400 keV, whereas the dominant neutron energy of the PHWR is 0.01 eV to 0.1 eV, as observed in Figure 4. When applying these dominant neutron energies to the cross-section plot in Figure 5, it can be seen that the samarium-149 neutron absorption cross-section in the PFBR is less than 1 barn, while the samarium-149 neutron absorption cross-section in the PHWR is around 1E+5 barns. This difference in neutron absorption cross-section in samarium-149 is hence the cause for the large PHWR to PFBR ratio (107) of samarium-150 concentration.

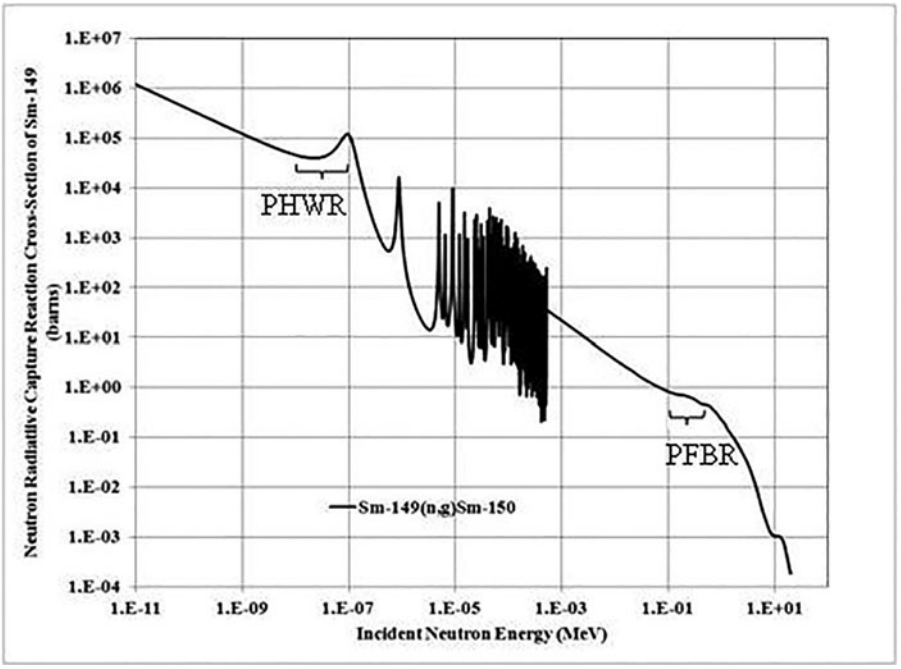

Figure 5: Plot of the neutron radiative capture cross-section for samarium-149.

Same Element Isotopic Ratios

Although the isotopes of an element behave very differently in nuclear reactions, they have very similar chemical properties. The fact that the isotopes of an element have similar chemical properties means they will behave similarly during PUREX chemical separation.[25] Therefore, select isotope ratios of the same element may infer details of the reactor system while being independent from the chemical separation process used. Table 6 gives ratios of the mass of isotopes of the same element present in weapons-grade plutonium produced in a PFBR and PHWR. The values for the isotope ratios found in PHWR plutonium are then divided by the PFBR values to represent the reactor dependency of the isotope of the same element ratios.

The cesium-137 to cesium-134 ratio has an observable difference while the ratios of samarium-150 to samarium-154 and plutonium-242 to plutonium-239 both contain more than an order-of-magnitude difference between the PFBR and PHWR. These three ratios could be measured by gamma spectroscopy, mass spectrometry, and alpha spectroscopy, respectively. It is therefore possible to deduce information about the producing reactor system from isotope ratios of the same element. The benefit of such ratios is that assumptions are not necessary to determine which chemical separation process was used and applying the appropriate decontamination factors. It can be inferred

Table 3: Selected isotopes per kilogram of PUREX processed plutonium from prototype fast breeder reactor radial blanket fuel.

Candidate Isotope	Expected mass (g) per 1 kg plutonium with DF of 10^6	Expected activity (Ci) per 1 kg plutonium with DF of 10^6
	Prompt Gamma	
Cesium-137	3.29E-06	2.86E-04
Cerium-144	1.14E-06	3.62E-03
	Alpha	
Plutonium-239	9.80E+02	6.08E+01
Plutonium-242	2.92E-03	1.15E-05
	Other Gamma	
Cesium-134	4.57E-08	5.92E-05
Antimony-125	2.73E-08	2.86E-05
Europium-154	1.54E-08	4.17E-6
	Mass Spectrometry	
Rubidium-85	1.87E-07	Stable Isotope
Strontium-90	1.12E-06	1.58E-04
Neodymium-148	1.04E-06	Stable Isotope
Promethium-147	9.59E-07	8.89E-04
Samarium-150	6.23E-08	Stable Isotope

Table 4: Selected isotopes per kilogram of PUREX processed plutonium from low-burn-up pressurized heavy water reactor fuel.

	Burn-up 1 MWd/kg		Burn-up 2 MWd/kg	
Candidate Isotope	Expected mass (g) per 1 kg plutonium with DF of 10^6	Expected activity (Ci) per 1 kg plutonium with DF of 10^6	Expected mass (g) per 1 kg plutonium with DF of 10^6	Expected activity (Ci) per 1 kg plutonium with DF of 10^6
		Prompt Gamma		
Cesium-137	4.23E-05	3.68E-03	4.46E-05	4.04E-03
Cerium-144	3.22E-05	1.03E-01	3.33E-05	1.06E-01
		Delayed Alpha		
Plutonium-239	9.60E+02	5.95E+01	9.15E+02	5.69E+01
Plutonium-242	5.60E-02	2.21E-04	4.29E-01	1.70E-03
		Other Gamma		
Cesium-134	1.30E-07	1.68E-04	3.92E-07	5.07E-04
Antimony-125	2.37E-07	2.48E-04	2.92E-07	3.06E-04
Europium-154	5.22E-08	1.41E-05	1.26E-07	3.40E-05
		Mass Spectrometry		
Rubidium-85	3.74E-06	Stable Isotope	3.89E-06	Stable Isotope
Strontium-90	2.43E-05	3.43E-03	2.52E-05	3.56E-03
Neodymium-148	1.31E-05	Stable Isotope	1.46E-05	Stable Isotope
Promethium-147	1.48E-05	1.38E-02	1.52E-05	1.41E-02
Samarium-150	6.67E-06	Stable Isotope	8.24E-06	Stable Isotope

Table 5: Reactor dependency of selected isotope ratios for pressurized heavy water reactor (PHWR) and fast breeder reactor (PFBR).

Candidate Isotope	Ratio of expected mass PHWR/PFBR
Prompt Gamma	
Cesium-137	12.86
Cerium-144	28.24
Delayed Alpha	
Plutonium-239	0.98
Plutonium-242	19.17
Other Gamma	
Cesium-134	2.84
Antimony-125	8.68
Europium-154	3.39
Mass Spectrometry	
Rubidium-85	20.00
Strontium-90	21.69
Neodymium-148	12.59
Promethium-147	15.43
Samarium-150	107.06

from Tables 5 and 6 that either the fission product to plutonium ratios (Table 5) or isotopic ratios of same element (Table 6) are independently capable in discriminating between weapons-grade plutonium produced from PHWR and PFBR. However, for any forensics analysis it is desirable to confirm the attributions using multiple and diverse signatures.

Stochastic Uncertainty

The MCNPX code used to simulate core operations is based on the principles of statistical stochastic methods for solving the Boltzmann radiation transport equation. Because of the stochastic nature of the solution method, the burn-up simulations were repeated by altering the sampling procedures to estimate the stochastic uncertainty (random error) associated with the predicted values of fission product and plutonium isotope concentrations. Burn-up

Table 6: Mass ratios for isotopes of the same element, pressurized heavy water reactor (PHWR) and fast breeder reactor (PFBR).

Isotope Ratio	Ratio of Isotope Mass from PHWR	Ratio of Isotope Mass from PFBR	Reactor Comparison PHWR/PFBR
$^{137}Cs/^{134}Cs$	3.25E+02	8.00E+01	4.07
$^{144}Ce/^{142}Ce$	8.04E-01	4.18E-01	1.92
$^{150}Sm/^{154}Sm$	8.88E+00	5.04E-01	17.6
$^{242}Pu/^{239}Pu$	5.83E-05	2.89E-06	20.2

simulations were repeated by changing the random seed number (a method used in MCNP to change the stochastic sampling procedures) and for each simulation, isotope concentrations were estimated. The average mass (μ) and one sigma standard deviation (σ) value for each isotope was calculated from the results of nine independent simulations. The relative random error (σ/μ) was obtained for the selected isotopes and found to be insignificant for all cases, with a random error of 0.37 percent or less for each isotope. This small random error indicates that the differences in the isotopic compositions seen from the reactor simulations are not due to the Monte Carlo method's random behavior.

The possibility exists for other sources of error in the model simulations that can affect the isotopic results. Monte Carlo methods have two types of uncertainties: random and systematic. The systematic uncertainty is associated with how close to reality the model is. We are assuming the systematic uncertainty is small.

CONCLUSIONS

Detailed computational models of the 500-MWe PFBR and 220-MWe PHWR cores were developed and fuel burn-up simulations performed using the radiation transport code, MCNPX. The objective of the reactor core burn-up simulations was to estimate the amount of plutonium and fission product isotopes contained in fuel discharged at low burn-up levels. The PFBR simulated normal operation with the radial blanket reaching a burn-up level around 1 MWd/kg. The PHWR models simulated the intentional discharge of fuel bundles at low burn-up levels of 1 MWd/kg and 2 MWd/kg. The plutonium isotopic composition bred in the fuel for these low burn-up cases would be that of weapons-grade plutonium and the objective of the study was to propose trace fission product to plutonium ratios and also same element isotope ratios (to avoid the uncertainty posed by varying decontamination factor) in separated weapons-grade plutonium for nuclear forensics purposes.

Simulation results for both reactors showed that at the low fuel burn-up levels, plutonium composition was that of weapons-grade with significant quantities of plutonium being produced.

After chemical separation of the plutonium from reprocessing operations, trace amounts of fission products will remain with the separated plutonium. Some fission products appeared to be good indicators of the type of reactor used for plutonium production. These candidate fission products were sorted into four groups according to the most suitable analytical technique for their respective quantification (rapid gamma-radiation measurement, delayed alpha-radiation measurement, slow gamma-radiation measurement, and mass spectrometry). The four group trace fission product to plutonium ratios (after applying a PUREX decontamination factor of 10^6) were estimated will be useful

for nuclear forensics purposes in the event of plutonium material interdiction because some of the ratios between the thermal reactor to fast reactor produced weapons-grade plutonium were significant and measurable. These isotopic ratios obtained for the fast PFBR and thermal PHWR were compared at the same fuel burn-up level. The comparisons showed that in case of an interdiction, the suite of selected isotopic ratios can attribute separated weapons-grade plutonium to a PHWR or PFBR. This is due to the fact that only two types of reactors could be compared in this study. However, in general the selected isotopic ratios are such that it can be applied to plutonium attribution to a fast or thermal neutron source reactor system.

Isotopic ratios of the same element were explored. The ratios of cesium-137 to cesium-134, samarium-150 to samarium-154 and plutonium-242 to plutonium-239 show that such ratios may lead to attribution of a source reactor system, while being independent of the chemical separation process used for plutonium separation.

Uncertainty estimates were made for the predicted concentrations of isotopes in the burned fuel because of the stochastic nature of MCNPX code simulations. The random errors estimated for the isotope concentrations were insignificant and indicated that the differences in the isotopic compositions between reactor simulations were not due to the random behavior of the Monte Carlo method.

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