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QUANTIFYING THE PASSIVE GAMMA SIGNAL FROM SPENT NUCLEAR FUEL IN SUPPORT OF DETERMINING THE PLUTONIUM CONTENT IN SPENT NUCLEAR FUEL WITH NON DESTRUCTIVE ASSAY

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ABSTRACT

The objective of safeguarding nuclear material is to deter diversions of significant quantities of nuclear materials by timely monitoring and detection. There are a variety of motivations for quantifying plutonium in spent fuel (SF), by means of nondestructive assay (NDA), in order to meet this goal. These motivations include the following: strengthening the capabilities of the International Atomic Energy Agencies ability to safeguard nuclear facilities, shipper/receiver difference, input accountability at reprocessing facilities and burnup credit at repositories. Many NDA techniques exist for measuring signatures from SF; however, no single NDA technique can, in isolation, quantify elemental plutonium in SF. A study has been undertaken to determine the best integrated combination of 13 NDA techniques for characterizing Pu mass in spent fuel. This paper focuses on the development of a passive gamma measurement system in support the spent fuel assay system. Gamma ray detection for fresh nuclear fuel focuses on gamma ray emissions that directly coincide with the actinides of interest to the assay. For example, the 186-keV gamma ray is generally used for ^{235}U assay and the 384-keV complex is generally used for assaying plutonium. In spent nuclear fuel, these signatures cannot be detected as the Compton continuum created from the fission products dominates the signal in this energy range. For SF, the measured gamma signatures from key fission products (^{134}Cs, ^{137}Cs, ^{154}Eu) are used to ascertain burnup, cooling time, and fissile content information. In this paper the Monte Carlo modeling set-up for a passive gamma spent fuel assay system will be described. The set-up of the system includes a germanium detector and an ion chamber, and will be used to gain passive gamma information that will be integrated into a system for determining Pu in SF. The passive gamma signal will be determined from a library of ~100 assemblies that have been created to examine the capability of all 13 NDA techniques. Presented in this paper is a description of the passive gamma monitoring instrument, explanation of the work completed thus far involving the source set up methodology and the design optimization process, details of key fission product ratios of interest, limitations and key strengths of the measurement technique, and considerations for integrating this technique with other NDA techniques in order to develop a complete spent fuel assay strategy.

INTRODUCTION

The objective of safeguarding nuclear material is to deter diversions of significant quantities of nuclear materials by timely monitoring and detection. [1] In order to meet this objective, the International Atomic Energy Agency (IAEA) inspector is tasked with accounting for all significant quantities of nuclear material through the various components of the nuclear fuel cycle. [2] These components include: enrichment, fuel fabrication, reactor operation, and spent fuel (SF) disposal (i.e. on site storage, repository storage, or reprocessing). To deter diversion through timely detection, assay strategies must be developed that are capable of quantifying the significant quantity

of interest in its various physical forms. For example, to determine plutonium content in an input accountability tank, and inspector may use the combination of the measured gross neutron count rate and knowledge of the ^{244}Cm/Pu-total ratio to determine total plutonium content in the tank. [3] The success of this assay strategy therefore becomes limited by the ability to assume that the tank is truly homogeneous, and that the sampled ^{244}Cm/Pu-total ratio may be extrapolated to the full volume of the tank. In another example, at a gaseous centrifuge enrichment facility, an inspector might analyze gross neutron counts, in UF_6 gas, resulting from the ^{234}U alpha decay and ^{19}F(alpha, n)^{22}Na reaction in order to determine ^{235}U enrichment. [4] The measurement success is limited to the understanding of how the signal becomes affected by material distribution in the tank. Therefore depending upon the form of the quantity of interest various limitations will have to be considered when developing an ultimate assessment strategy.

From the time the reactor pressure vessel lid has been opened for a refueling to the time of reprocessing or ultimate fuel storage, SF offers a diversion pathway of significant quantities of plutonium. In terms of plutonium content, SF is heterogeneous containing varied actinide content from pin-to-pin (from power peaking due to boundary leakage and water rods) as well as actinide gradients across a fuel pin (from low energy neutron resonances, such as the 6.67 eV U-238 resonance, resulting in short neutron mean free paths). [5]

Destructive analysis (DA) is capable of extrapolating plutonium content in a small sample to determine the content in the entire SF assembly; however, DA is both costly and time consuming, involving on-site laboratory space, incapable of determining fuel pin diversion by way of pin substitution, and therefore not a practical technique for assaying plutonium content in spent fuel at a reprocessing facility or repository intake.

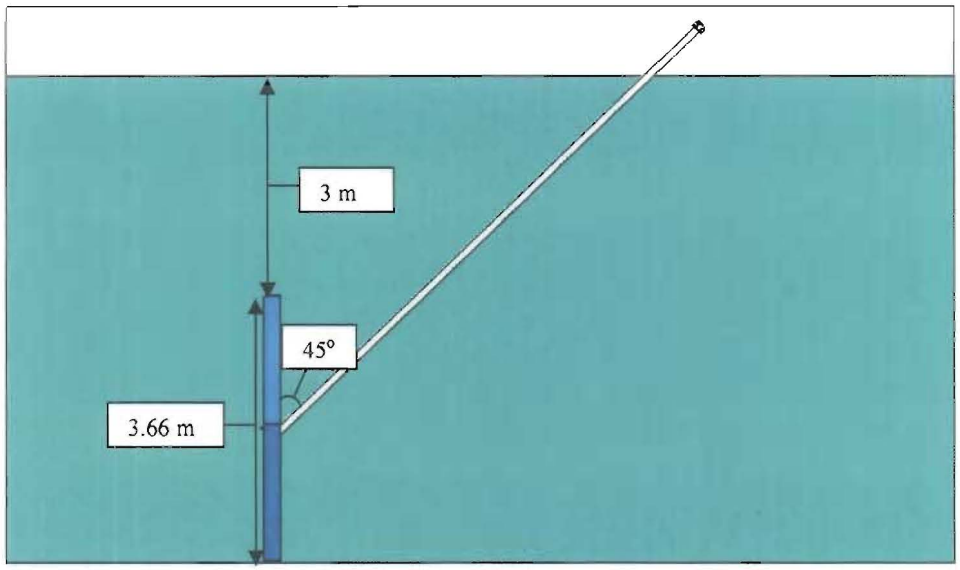
Nondestructive assay (NDA) offers a cost efficient, timely, standardized method of SF assay capable of determining pin diversion and therefore capable of meeting the objectives of nuclear safeguards for SF. [6] In fact, there are a variety of motivations for quantifying plutonium in SF, by means of NDA, in order to meet this goal. These motivations include: (1) strengthening the capabilities of the International Atomic Energy Agency ability to safeguard nuclear facilities; (2) shipper/receiver difference; (3) input accountability at reprocessing facilities; (4) burnup credit at repositories; and (5) continuity of knowledge at the SF storage site. Many NDA techniques exist for measuring signatures from SF; however, no single NDA technique can, in isolation, quantify elemental plutonium in SF. A study has been undertaken to determine the best integrated combination of 13 NDA techniques for characterizing Pu mass in spent fuel. [7] The techniques include: Delayed Gamma, Delayed Neutrons, Differential Die-Away, Differential Die-Away Self-Interrogation, Lead Slowing Down Spectrometry, Neutron Multiplicity, Nuclear Resonance Fluorescence, Passive Prompt Gamma, Passive Neutron Albedo Reactivity, Self-Integration Neutron Resonance Densitometry, Total Neutron (Gross Neutron), X-Ray Fluorescence, and ^{252}Cf Interrogation with Prompt Neutron Detection. These techniques rely on the detection of either gamma or neutron emission from either induced or passive events, and therefore each technique has advantages and disadvantages towards meeting the objective of plutonium assay in SF. This paper focuses on the initial development of a passive gamma measurement system in support the spent fuel assay system. Presented in this paper is a description of the passive gamma monitoring

instrument, explanation of the work completed thus far involving the source set up methodology and the design optimization process, details of key fission product ratios of interest, limitations and key strengths of the measurement technique, and considerations for integrating this technique with other NDA techniques in order to develop a complete spent fuel assay strategy.

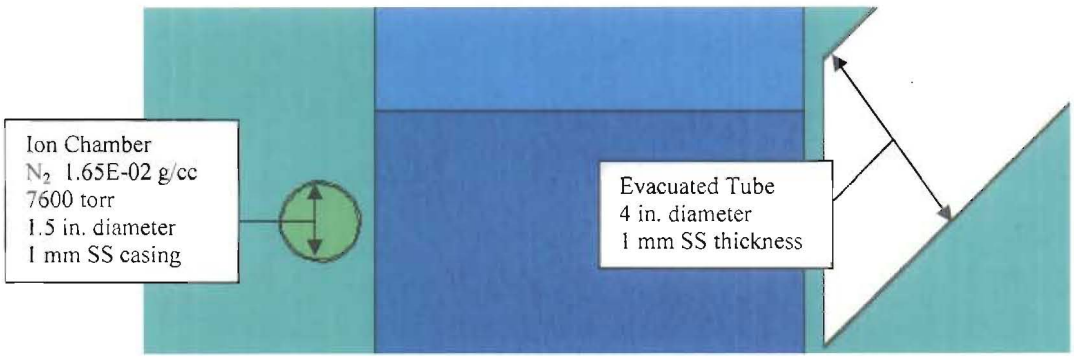
PASSIVE GAMMA SYSTEM FOR SPENT FUEL ASSAY

Gamma ray detection for fresh nuclear fuel focuses on gamma ray emissions that directly coincide with the actinides of interest to the assay. For example, the 186-keV gamma ray is generally used for ^{235}U assay and the 384-keV complex is generally used for assaying plutonium. In SF, most fission product signatures range from 600 keV to a few MeV and therefore cause the low energy ^{235}U and plutonium signatures to fade into the noise as the Compton continuum created from the fission products saturates the signal in the low energy regime. For SF, the measured gamma signatures from key fission products (^{134}Cs, ^{137}Cs, ^{154}Eu) have been used to ascertain burnup, cooling time, and fissile content information. [8-10] For example, the ratio of the ^{134}Cs (795.8 keV) to ^{137}Cs (661.7 keV) gammas scales with fuel assembly burnup while the ratio of ^{134}Cs (795.8 keV) to ^{154}Eu (1274.4 keV) can be used to possibly determine U/Pu ratios. The two major limitation of utilizing these signatures in the context of SF measurement are: (1) attenuation of the signal across the assembly; and (2) variation of the ratio as a function of cooling time due to varying half-lives. Uranium oxide significantly attenuates the gamma signal across the bundle, and thus a passive gamma instrument could never be used by itself to determine if a diversion had taken place. Nonetheless, a passive gamma measurement will give useful information from the SF periphery that can be used to extrapolate actinide buildup across the whole assembly. Therefore one strategy would be to use a passive gamma instrument to estimate burnup and couple the instrument with a neutron measurement estimating burnup from the singles count rate in order to see if the same burnup estimate is achieved with both instruments. If discrepancy existed between the two instruments, then the inspector could infer that diversion has taken place. Of course, the varying half-lives of the fission product signatures and therefore variation of the signal ratios as a function of cooling time would limit the effectiveness of the technique. However, also employing a gross gamma counting method along with a spectra calculation would help mitigate these issues.

Figure 1a-b displays radial profiles of the passive gamma counting system. The passive gamma counting system is composed of an ion chamber (for gross gamma counting) and an evacuated collimator attached to a germanium detector (displayed in the figure is just the front end of the germanium crystal). The ion chamber is composed of N_2 fill gas at a pressure of 7600 torr, and has a 5.89 in. length and 1.5 in. diameter with a 1 mm thick stainless steel casing. The evacuated collimator has a 4 in. diameter and is ~7.5 m long in order to account for the height of the liquid nitrogen cooling dewar (not displayed in figure 1a). Both the evacuated collimator and ion chamber are placed 1 cm from the SF periphery. The germanium crystal is 7.5 cm. diameter by 7.5 cm length. A 10 mm vacuum gap exists between the sides of the crystal and the 1 mm thick aluminum casing while a 5mm gap exists between the top of the crystal and the aluminum window. For simplicity in the initial design, only 2.5 cm of lead shielding surrounds the aluminum casing.



(a)



(b)

Figure 1. Radial profile of the passive gamma counting system: (a) large scale system orientation; (b) zoomed in profile displaying ion chamber and evacuated tube for germanium counting.

SOURCE SETUP AND DESIGN OPTIMIZATION CHALLENGES

Since the radiation signature of SF depends upon the reactor operating characteristics, assembly type, initial enrichment, burnup, cooling time and diversion scenario, a Monte Carlo linked depletion SF library was developed for assessing each of the 13 NDA techniques. [5] A diversion scenario library was also generated to test out the sensitivity of each instrument to various amounts of diversion from various locations. [11] The literature states that nearly 92% of the ^{137}Cs signal and 72% of the ^{154}Eu signal originates in the outer three rows of fuel pins for a 15 X 15 SF assembly (the higher energy 1.2744 MeV ^{154}Eu gamma is more penetrating than the 0.6616 MeV ^{137}Cs gamma); therefore the passive gamma system is not expected to be effective for measuring diversion by itself. [12] Nonetheless understanding the change in signal versus the varied measurement conditions in the SF library for over ~100 assemblies will determine how this technique will best complement another technique.

The spent fuel library only contains masses of nuclides for which transport cross sections are available. The masses of all the other 3000 nuclides without transport cross sections are tracked in the depletion module; however, these nuclide masses are not reported in the output file. This limitation impairs the ability to generate the complete gross gamma signal at any given time step; however, for the cooling times examined for our counting purposes, most of the short lived radioactive nuclides have decayed into longer lived daughters containing long enough half-lives that transport cross sections have been developed for these nuclides. Therefore the issue of generating adequate gamma source terms at a given burnup and cooling is only limited by the ability to generate the expected gamma emissions from these longer lived radioactive fission products.

MCNPX 2.7.A possess a spontaneous photon emission feature capable of automatically generating the decay gamma lines from radioactive fission products. [13] The emission of each gamma is based on the combination of the probability of an isotope being created by a decay and the branching ratio of given gamma from a decay. The feature can automatically determine the activity of a given cell and also sample in multi-cell geometries based on the activity of a given series of cells. At first this capability seemed to be useful for generating the passive gamma source since the spent fuel model contains ~1000 emission regions; however, a few difficulties were encountered.

Most fission product decay chains are short and therefore only a few gammas at various branching energies are produced per fission product nuclide decay. A simple test using MCNPX 2.7.A with a multi-cell lattice cylinder source composed of ^{60}Co and ^{134}Cs contained within a water box was able to quickly create the decay gammas and transport them. The same test, however, had significantly longer run time when implementing actinides as a passive gamma source. Actinide decay chains are quite long with sometimes more than 15 decays before reaching a stable daughter. These long decay chains result in hundreds of possible branching gammas with most branching energies having low probability of emission. In fact, the decay gammas of the longer lived actinides have a lower probability of emission. In composing the cumulative distribution function (CDF) for sampling the decay gammas lines, the probability that a given gamma line is sampled is simply the product of the activity of a given isotope in a material and the branching ratio of a specific gamma line. In a decay chain, the probability of emission of decay gammas is therefore dependent upon the decay of the preceding species; therefore decay chains that start with a nuclide of low activity will have a low probability of sampling other gammas within that chain. Because MCNPX is trying to determine a single decay line for a single history, many particles must be simulated in order to adequately sample all emitted gammas of interest. Furthermore, there is significant computational expense associated with generating gammas from large decay chains, such as actinide decays, which result in a complex CDF containing many gamma lines in a decay chain that probably won't be sampled often. Therefore the current spontaneous photon capability in MCNPX 2.7.A was currently deemed incapable of practical implementation for generating a passive gamma source due to the amount of actinides in our passive gamma source and the extensive computational time wasted generating the CDF for low probability events.

Another method was selected for modeling the passive gamma source. This method will involve: (1) extracting the activity of each available radionuclide from the MCNPX Monte Carlo burnup

output file; (2) multiplying the activity of each available radionuclide by the branching ratio of each of the radio nuclides decay gamma to determine its contribution; (3) summing the contributions to determine, total and then quantify the nuclides that compose at least 1% of the signal; (4) generating an sdef card that is composed of separate emissions for each separate material sampling from only the nuclide decay lines that contribute at least 1% to the total emission. As of the time of this paper, the implementation of this method is still in progress.

KEY FISSION PRODUCT RATIOS OF INTEREST

The passive gamma system is set up to measure both gross gamma and spectroscopic information. At first thought, one might think it best to set up fission product monitors for the highest activity high energy gamma emitters as these emitters are attenuated less, and therefore may escape from deeper inside the SF assembly. However, not all of these emitters have long enough half-lives to be significant monitors for SF at longer cooling time and not all of these emitters has fission product yields that are beneficial for determining intrinsic properties of the SF. For example, ^{134}Cs has a thermal fission product yield that is similar in ^{235}U, ^{239}Pu, and ^{241}Pu fission and therefore acts as a great indicator of total fission energy and can be used to determine burnup (thermal fission yields for ^{134}Cs from ^{235}U, ^{239}Pu, and ^{241}Pu fission are 0.067, 0.070, 0.067). [14] Isotopes like ^{154}Eu have significantly different fission product yields for ^{235}U, ^{239}Pu, and ^{241}Pu fission and can be used as indicators for ratios of fissile nuclides (thermal fission yields for ^{154}Eu from ^{235}U, ^{239}Pu, and ^{241}Pu fission are 1.53E-03, 3.61E-03, and 5.41E-03). [14] Examining the combinations of these emitters can give information regarding burnup and expected fissile nuclide ratios. Furthermore, examining ratios of the nuclides will help to eliminate uncertainty in geometric arrangement of the detector and therefore only requires that relative detector efficiency be known. [15]

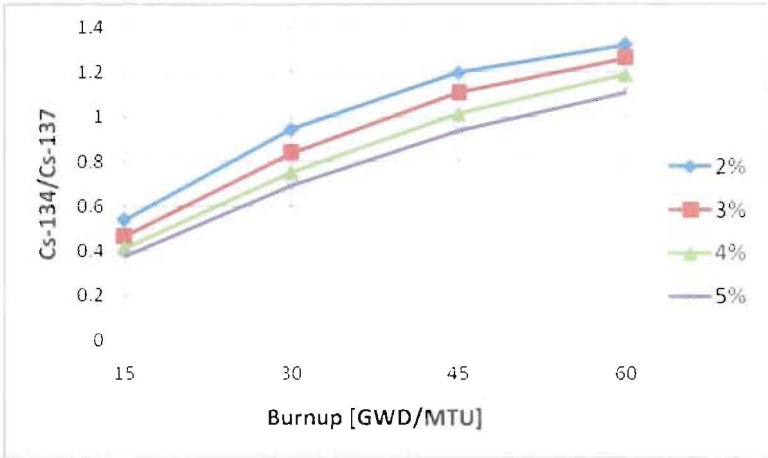
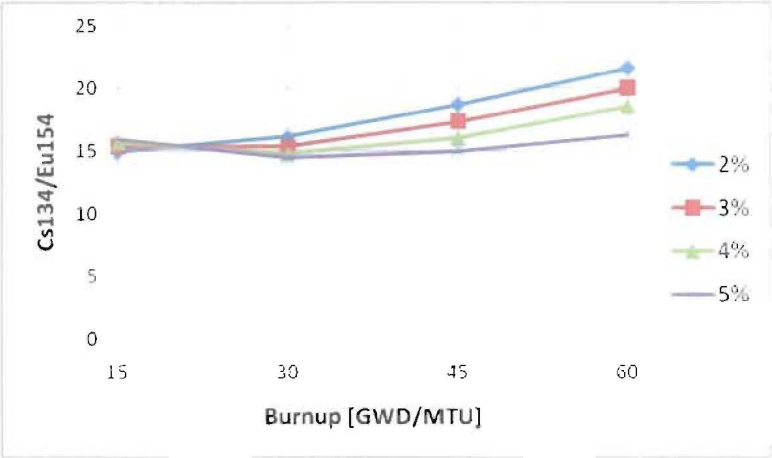


Figure 2 ^{134}Cs/^{137}Cs activity ratio as a function of burnup for various enrichments 2 year cooled.

Figure 2 displays the ^{134}Cs/^{137}Cs ratio as a function of burnup for various enrichments. The ^{134}Cs/^{137}Cs ratio increases almost linearly with burnup for each enrichment (slight parabolic behavior is evident above 45 GWD/MTU). Since power is proportional to macroscopic fission cross section multiplied by the flux, for a decrease in macroscopic fission cross section, the flux must increase in order to maintain constant power. Therefore for the same equivalent power, a lower enriched fuel assembly will have a higher flux. The creation of both ^{134}Cs (created by

$^{133}Cs(n,\gamma)^{134}Cs$) and ^{154}Eu (created by $^{153}Eu(n,\gamma)^{154}Eu$) rely on a neutron capture event and therefore the buildup of both of these nuclides is directly proportional to the fluence magnitude as well as the fission rate (^{137}Cs is created directly as a fission product and there its buildup is only related to the exact fission rate). The flux magnitude percent difference between 2% and 5% enriched is an order of magnitude greater than the difference in fission rate. Though the creation of ^{134}Cs and ^{154}Eu requires a fission event to occur, the buildup of these nuclides as a function of enrichment difference is more dependent upon flux level and thus the lower enriched fuel assembly generates more ^{134}Cs and ^{154}Eu for an equivalent power level. Therefore measuring the quantity of $^{134}Cs/^{137}Cs$ by itself can be used as a diagnostic for burnup if the initial enrichment is known (and vice versa).



$^{134}Cs/^{154}Eu$ activity ratio as a function of burnup for various enrichments 2 year cooled.

The fission yield of ^{154}Eu varies significantly as a function of fissioning species. Therefore because ^{134}Cs does not vary significantly as function of fissioning species, it is thought that the ratio of $^{134}Cs/^{154}Eu$ can be used to determine the expected amount of plutonium in the system. Figure 3 displays the $^{134}Cs/^{154}Eu$ ratio as a function of burnup for various enrichments. At 15 GWD/MTU, ^{235}U fission contributes at a higher percentage for higher enrichment, therefore less ^{154}Eu is created at this burnup for higher enrichments which increases the $^{134}Cs/^{154}Eu$ ratio. For 60 GWD/MTU burnup, the ^{134}Eu content increases while the ^{134}Cs content decreases with increasing enrichment suggesting that more plutonium is being burned at higher enrichments and 60 GWD/MTU burnup. The significant decrease in ^{134}Cs as function of enrichment still dominates the ratio and therefore $^{134}Cs/^{154}Eu$ may not be as an effective passive plutonium monitor as it would be for active gamma counting due to the amount of dominating time integrated fissions of ^{235}U. However, because the signal behaves differently than the $^{134}Cs/^{137}Cs$ activity ratio, it may be possible to use the combination of both signals to ascertain direct information regarding burnup, initial enrichment, and plutonium content.

CONCLUSIONS

A passive gamma counting system has been proposed as a component to an integrated spent fuel assessment strategy. The instrumentation includes both an ion chamber for gross gamma counting and germanium detector for spectroscopic information. As low energy gamma emission from

actinide decay is expected to be masked by the Compton continuum from the scatter of high energy gammas from fission product emission, this system will focus on collecting fission product signal ratios in order to ascertain information regarding initial enrichment and burnup. Though difficulties were encountered using new automated features in MCNPX 2.7.A, a methodology has been mapped out for developing the passive gamma emission spectra for the ~100 assemblies available in the spent fuel library used to examine the capability of all 13 NDA techniques. Activity ratios of key fission products have been presented and the results suggest that these ratios are able to correlate with burnup and initial enrichment.

A passive gamma system will not be capable of acquiring signal information from the SF interior region, and therefore will be ineffective in assessing SF inner region diversion. None of the instruments is expected to be a magic bullet for plutonium assay in spent fuel. However, a passive gamma system will be capable of achieving an independent measurement of burnup on the fuel assembly exterior, which can be used to extrapolate across the entire SF assembly. Though neutrons are less attenuated and therefore penetrate out from the SF interior region, the signal is dependent upon initial enrichment, burnup, and diversion scenario. In some cases these interdependencies cause overlap, therefore making it difficult to pinpoint a "state" of the assembly. A passive gamma system will complement this type of measurement by providing extrapolated information based on a signal from a portion of the SF assembly that is expected to be unperturbed. Then the combination of these instruments will be able to determine the SF "state," by trying to match expected burnups from each type of measurement.

Future work for this study will involve: (1) completing the developing of the passive gamma emission source; (2) characterizing the passive gamma emission spectra versus burnup, initial enrichment, diversion scenario and cooling time; (3) further developing a methodology for integrating a gamma counting technique. It is important to realize that this study did not address cesium migration as function of temperature gradient and fuel assembly burnup. Cesium migrates across a thermal gradient from hot to cold regions and therefore is thought to affect using this nuclide as a burnup indicator; however, using the ratios of nuclides of the cesium emission signal is thought to minimize the impact of this effect. Passive gamma signal detection has the advantage that no active source is required; however, the major disadvantages are: (1) the signal is highly attenuated from the SF interior region; (2) the signal is dependent upon the time integration of interactions incurred during reactor operation, and therefore more limiting than a direct active measurement. Therefore this technique may or may not be part of the ultimate SF plutonium assay strategy, and future analysis of other active systems will be required to determine if a passive gamma detection system will be part of the ultimate plutonium assessment strategy.

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