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A Monte Carlo Based Spent Fuel Analysis Safeguards Strategy Assessment

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Abstract –*Safeguarding nuclear material involves the detection of diversions of significant quantities of nuclear materials, and the deterrence of such diversions by the risk of early detection. There are a variety of motivations for quantifying plutonium in spent fuel assemblies by means of nondestructive assay (NDA) including the following: strengthening the capabilities of the International Atomic Energy Agencies ability to safeguards nuclear facilities, shipper/receiver difference, input accountability at reprocessing facilities and burnup credit at repositories. Many NDA techniques exist for measuring signatures from spent fuel; however, no single NDA technique can, in isolation, quantify elemental plutonium and other actinides of interest in spent fuel. A study has been undertaken to determine the best integrated combination of cost effective techniques for quantifying plutonium mass in spent fuel for nuclear safeguards.*

A standardized assessment process was developed to compare the effective merits and faults of 12 different detection techniques in order to integrate a few techniques and to down-select among the techniques in preparation for experiments. The process involves generating a basis burnup/enrichment/cooling time dependent spent fuel assembly library, creating diversion scenarios, developing detector models and quantifying the capability of each NDA technique. Because hundreds of input and output files must be managed in the couplings of data transitions for the different facets of the assessment process, a graphical user interface (GUI) was development that automates the process. This GUI allows users to visually create diversion scenarios with varied replacement materials, and generate a MCNPX fixed source detector assessment input file. The end result of the assembly library assessment is to select a set of common source terms and diversion scenarios for quantifying the capability of each of the 12 NDA techniques.

We present here the generalized assessment process, the techniques employed to automate the coupled facets of the assessment process, and the standard burnup/enrichment/cooling time dependent spent fuel assembly library. We also clearly define the diversion scenarios that will be analyzed during the standardized assessments. Though this study is currently limited to generic PWR assemblies, it is expected that the results of the assessment will yield an adequate spent fuel analysis strategy knowledge that will help the down-select process for other reactor types.

I. INTRODUCTION

Safeguarding nuclear material involves the detection of diversions of significant quantities of nuclear materials, and the deterrence of such diversions by the risk of early detection.[1] To meet this objective, the International Atomic Energy Agency (IAEA) inspector is tasked with determining if diversions of significant quantities have taken place, and preventing these diversions, by accounting for all significant quantities of nuclear material during various components of the nuclear fuel cycle.[2] The components of most interest include: enrichment, fuel fabrication, reactor power production, and spent fuel disposal (onsite storage, repository storage or reprocessing).[3]

For these components of the nuclear fuel cycle, significant quantities of nuclear materials will exist in various forms. 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.[4] At a spent fuel reprocessing facility, an inspector might be counting ^{244}Cm neutrons in a spent fuel waste stream and using the ^{244}Cm/Pu-total ratio to determine total plutonium content.[5] Therefore the material of interest will vary depending upon the component of the nuclear fuel cycle.

From the time the reactor is opened for a refueling to the time of reprocessing or ultimate off site repository disposal, spent nuclear fuel assemblies, offers an opportunity for the diversion of significant quantities of nuclear material in the form of plutonium. Therefore during these components of the nuclear fuel cycle, the inspector is tasked with accounting for significant quantities of plutonium within a nuclear fuel assembly. There exist many methods for accounting for spent fuel tampering including: fuel assembly ID verification, tags and seals, destructive analysis (DA), and nondestructive assay (NDA).[6] The first three methods contain caveats of interest: (1) fuel assembly ID verification with operator logs is subject to operator fraud; (2) tags and seals may be faked or altered; (3) destructive analysis, though thorough, may be time and cost prohibitive and further result in the destruction of possibly useful material. NDA offers a cost efficient method to assay the abundance of spent fuel assemblies at any given site to meet the objectives of nuclear safeguards. There are also a variety of other motivations for quantifying plutonium in spent fuel assemblies by means of NDA including the following: shipper/receiver difference, input accountability at reprocessing facilities, optimizing the location of partially burned assemblies in the core and burnup credit at repositories.

Historically, Cerenkov glow detection has been used as a strategy for qualitatively determining spent fuel tampering.[7] This strategy relies upon the IAEA inspector visually assessing the Cerenkov glow created by high-speed electrons, generated from the intense radioactivity of the fuel.[6] The inspector can, if sufficient measurements are taken, ascertain qualitative information regarding assembly burnup, cooling time, and tampering based on the characteristics of the Cerenkov glow. However, because the measurement technique depends on visual inspection with inspector interpretation (for digital systems with image subtraction the user must still position a hand-held instrument), different levels of experience, from inspector to inspector, or even measurement time to next measurement time, may be insufficient for the future of nuclear safeguards inspections. NDA techniques, which give quantitative isotopic analysis, are most effective for standardized safeguards measurement at more components of the nuclear fuel cycle because the techniques can quantify plutonium and do not rely on indirect indicators of the presence of plutonium.

NDA strategies are currently utilized at fuel fabrication facilities for successful fuel specification testing. For example, the coincidence fuel rod scanner is an active neutron coincidence counter used to determine total fissile loading. The counting system uses an active source, such as ^{252}Cf, to interrogate a fuel pin in order to induce fission events. Those fission events create multiplicities of neutrons that are later detected as coincident counts. Knowing the moments of the multiplicity distribution, one can calculate the concentration of a fissile species. If the fuel content contains only one fissile species, then the counter is used to determine the content of that species. Unfortunately, this technique becomes more complicated for spent fuel isotopic assay as many varied fissile nuclides exist in a given spent fuel rod, and therefore isolation of a specific fissile nuclide requires further correction and deconvolution. Another example of a UO_2 fuel specification complicated by spent fuel measurement is passive gamma assay. For fresh fuel, using attenuation correction, one can assay ^{235}U content using the 186-keV gamma ray, and one can also assay plutonium using the 386-keV complex. For specification testing one can usually safely assume a homogenously mixed pellet; therefore leading to an effective attenuation correction factor. Spent fuel emits many high energy gamma rays resulting from fission product decay. The Compton continuum of these high energy gammas dominates the low energy signal resulting in the keV signatures, of ^{235}U and the plutonium complex, fading into the noise; furthermore, due to the inhomogeneous burnup of the fuel pin, the gamma emission rate is not uniform further complicating attenuation correction. Therefore individual fissile nuclide assay of spent fuel using NDA will be complicated by: (1) the inhomogeneous nature of the fuel particle emissions; (2) the various amounts of fissile nuclides; (3) fission product gamma emission.

Many NDA techniques exist for measuring signatures from spent fuel; however, no single NDA technique can, in isolation, quantify elemental plutonium and other actinides of interest in spent fuel.[7,8,9] A study has been undertaken to determine the best integrated combination of cost effective techniques for characterizing Pu mass in spent fuel for nuclear safeguards.[9] The study will seek to analyze the merits and faults of 12 different NDA techniques in order to down-select to an integrated spent fuel assay strategy for nuclear safeguards. The 12 NDA techniques being researched include the following: Delayed Gamma, Delayed Neutrons, Differential Die-Away, 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, ^{252}Cf Interrogation with Prompt Neutron Detection.[9] Each technique obtains information from either a gamma or neutron signal, measuring either gross counts or multiplicities, from either active induced or passive signal analysis, and thus each technique will have both advantages and limitations for assaying plutonium content in spent nuclear fuel.

A standardized assessment process was developed to compare the effective merits and faults of 12 different detection techniques in order to integrate a few techniques and to down-select among the techniques in preparation for experiments. The process involves generating a basis burnup/enrichment/cooling time dependent spent fuel assembly library, determining diversion scenarios, developing detector models and quantifying the capability of each NDA technique. This paper first presents the generalized assessment process briefly focusing on the software employed to automate the assessment's coupled facets and the standardized burnup/enrichment/cooling time dependent spent fuel library (SFL) used as the source term for comparison. This paper clearly defines the mechanisms and considerations for selecting the diversion scenarios and lists the final selected diversion maps used for comparison.

II. AUTOMATING THE INTEGRATED PROCESS

The generalized assessment for down-selecting the 12 NDA techniques involves consideration of many different types of interaction of physics concern. For example, the spatial dependence of the burnup results in a spatial gradient of the nuclide buildup. The spatial gradient of the nuclide buildup further affects the emission signal; therefore spatial self-shielding is of significant consideration for some of the NDA techniques Multi-particle physics concerns are also important for assessing interference of different particle types (i.e. gamma energy deposition in a neutron counter) for different types of NDA strategies. Most importantly, the code utilized for the assessment must be able to generate and report the quantities of interest. For example, one must be able to generate a passive source of spontaneous fission neutrons and be able to account for coincident detection and report the moments of the multiplicity distribution. Therefore MCNPX 2.7.A was chosen as the modeling tool for the assessment effort because of the code's ability to achieve high spatial resolution without concern for spatial self-shielding approaches, model multi-particle physics necessary for detector calculations, generate a spontaneous fission source, tally on time dependent quantities of interest (i.e. coincidence counting and multiplicity moments distributions), and minimize assumptions necessary for standardized code-to-code coupling.[10]

The generalized assessment for down-selecting among the 12 NDA techniques also involves many different types of calculations. For example, first a case matrix of spatially dependent burnup calculations is required in order to generate a SFL used as a source term for comparison of each NDA technique. In parallel to the development of the SFL, base detector models have to be engineered. Base diversion scenarios have to also be determined and developed. Finally, the expected performance of each NDA technique must be quantified for the cases of the source term library, diversion scenarios. Therefore more than a thousand, input files will be run in order to complete this assessment.

Because over a thousand input and output files must be managed in the couplings of data transitions for the different facets of the assessment process, a graphical user interface (GUI) was development that automates the process. The Burnup Automation MCNPX File Data Retrieval Tool (BAMF-DRT 1.0) allows users to visually create diversion scenarios with varied replacement materials, and generate a MCNPX fixed source detector assessment input file.

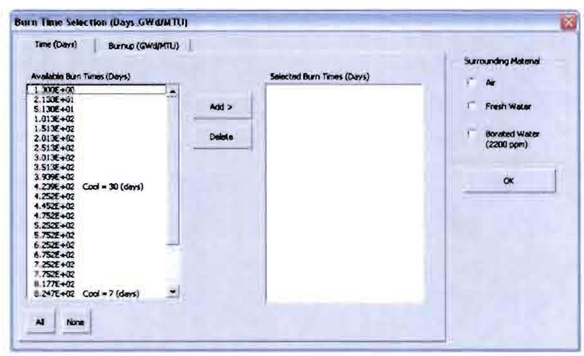

Fig. 1a. BAMF-DRT 1.0 Burnup Time Selection User Form.

BAMF-DRT 1.0 uses the VBA Excel platform to automate the coupling of MCNPX burnup output files, accruing information of importance, generating a fixed source assembly file for a given cooling time, combining a detector geometry and a fuel assembly file, and generating diversion assemblies through the use of visual user forms containing easy-to-use drop down menus and push button execution.[11] Figs. 1a-c display example BAMF-DRT 1.0 user forms. After a user selects an MCNPX burnup output file, the available burnups and cooling times, from which BAMF-DRT 1.0 is able to ascertain mass information, are loaded into the user form in Fig. 1a. The user may then select the matrix material (water or air being the most likely) and as many burnups and/or cooling times as they wish in order to generate assembly files containing burnup and/or cooling time dependent mass information. The user may then wish to "divert" fuel pins and replace the fuel material within those pins with diversion material. BAMF-

DRT 1.0 helps the user make these diversions through the use of a visual diversion map as displayed in Fig. 1b (in this picture guide tube universes are black). The user may then choose to highlight pins they wish to divert, click on the color of the diversion material they wish to use, and the diversion will be made automatically. Any type of diversion pattern can be made. For example, the user can divert an entire highlighted row and checkerboard pattern at the same time.

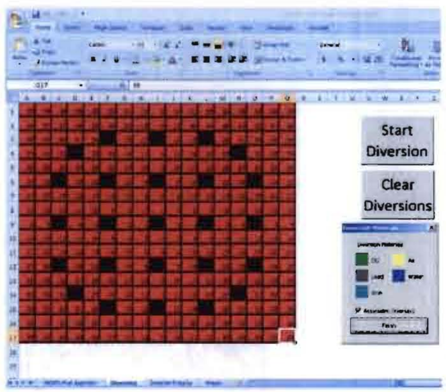

Fig. 1b. BAMF-DRT 1.0 Diversion Map.

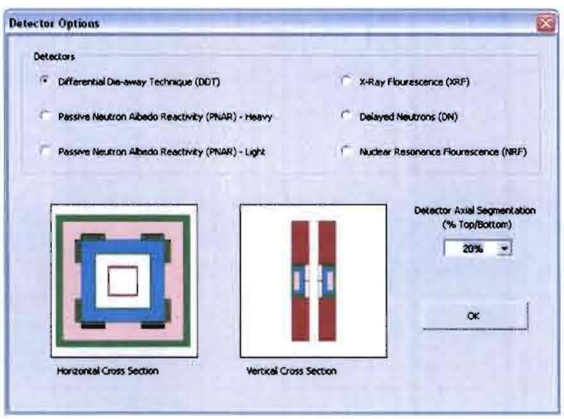

Fig. 1c. BAMF-DRT 1.0 Detector Options User Form.

After a diversion scenario is completed or if the user chooses to bypass generating a diversion, the user must then select a detector to combine with the fuel assembly file. Fig. 1c displays the easy to use detector option user form for selecting a detector. When the user clicks on a detector option in the "Detectors" frame, an axial and radial cross sectional view of the detector geometry appears on the user form. The user simply clicks the detector geometry they wish to use, presses the OK button and the fixed source input file is automatically generated. The BAMF-DRT 1.0 procedure is powerful automation tool that streamlines the assessment process by minimizing the transitioning of data from one calculation to another in order to maximize the time spent actually analyzing data.

III. SPENT FUEL LIBRARY

The radiation signature of a spent fuel assembly depends upon the reactor operating characteristics, assembly type, initial enrichment, burnup, cooling time, and diversion scenario. A base burnup/ enrichment/ cooling time dependent spent fuel library was developed based on a typical pressurized water reactor (PWR) spent fuel assembly in order to be used as a source term for comparison of the faults and merits of each NDA technique.

The MCNPX burnup source term library consists of a typical Westinghouse 17 X 17 PWR spent fuel assembly burned to varied burnups, with typical initial enrichments, consistent with the range of PWR spent fuel available today. The details of the fuel dimensions, cladding material, boron concentrations, and temperatures are detailed in Ref. 12. The fuel region is subdivided into four subregions in order to capture the spatial gradient of higher actinide buildup resulting from the short mean free path of neutrons around the large 6.67 eV resonance of ^{238}U (results in a significant gradient within the first 200 microns of the fuel pellet). Capturing this gradient is important for assessing X-ray fluorescence from which the signal primarily comes from the outer 200 microns of the fuel. Continuous energy Monte Carlo is an excellent candidate for spatial gradient burnup problems as the user may segment the problem geometry into many radial zones without the concerns of spatial self-shielding treatment. The depletion capability in MCNPX 2.7.A had already been successfully benchmarked for LWR calculations and therefore was a natural choice for generating the source term library.[13]

The library was generated burning an infinitely reflected fuel assembly with 2%, 3%, 4%, and 5% initial enrichment to 15, 30, 45, and 60 GWD/MTU, and having cooling times of 1 and 4 weeks, and 1, 2, 5, 20, and 80 years. First short burn steps were generated to account for initial Xe and Sm buildup, and then burn steps were generated that did not exceed ~2 GWD/MTU after Xe and Sm buildup. Each KCODE time step utilized 10000 particles per cycle, skipping the first 25 cycles for 155 cycles. Details on time durations, computational times, and compiler and message pass interface combinations are given in Ref. 12.

IV. DIVERSION SCENARIOS

The objective of nuclear safeguards is the timely detection of diversion of significant quantities of nuclear materials, and the deterrence of such diversion by the risk of early detection.[1] As previously discussed, significant quantities of nuclear material can be diverted at different steps in the nuclear fuel cycle (i.e. UF_6 gas during enrichment, fuel pins during reactor operation and spent fuel storage, and actinide waste streams during reprocessing). If a proliferator wished to divert nuclear material from a fuel assembly, the proliferator must either divert the entire assembly or divert fuel pins within the assembly. With proper continuity of knowledge and material accounting, diverting pins from an assembly is harder for an inspector to detect than the absence of an entire assembly. In diverting fuel pins, one can choose to either extract fuel pins from a fuel assembly leaving no replacement or extract fuel pins from an assembly inserting a dummy fuel pin containing diversion material.

Proliferators may choose a diversion material based on the limitations of the available diversion equipment. For example, if time is of concern, a proliferator may choose to load empty pins, water filled pins, or pins filled with iron pellets. If a proliferator wished to be more furtive, many other options for diversion materials based on the expected inspector measurements. For example, the manufacturing density of a UO_2 is ~10.45 g/cc; therefore one may expect a proliferator to use lead, which has a density of 11.72 g/cc, as a diversion material so that no noticeable change in assembly weight is detected. Within that same line thinking, if available, a proliferator may divert spent pins and replace with low enriched uranium (LEU) or depleted uranium (DU) pins. Certainly one can make the argument that a proliferator would probably not choose to select a diversion material that was itself also safeguarded, such as LEU, and therefore choose to divert with non safeguarded material such as DU. However, the fissile content in LEU offers a significant attribute to testing the limitations of a given techniques ability to assess plutonium diversion as the effective fissile content in LEU can be made to match the effective fissile content in a spent fuel pin. Since these types of limitations exist, these types of diversions need to be examined in order to develop an ultimate spent fuel assay strategy capable of timely detecting these types of diversion scenarios.

The best way to determine the most limiting pin diversion locations is by determining the importance of a given pin location to the detectable signal. Deterministically, one would think to accomplish this task by solving for the adjoint flux from the adjoint linear Boltzmann transport equation where the adjoint flux is the importance of fuel pin to a detection scenario. In MCNPX 2.7.A, one can calculate the contribution of a given particle to a given tally by use of the tally tagging feature. The tally tagging feature is capable of determining the uncollided source from a given cell to a given tally as well as determining the amount of a given reaction in a given cell that emits particles that contribute to a given tally.

Some of the 12 NDA techniques examined rely on photon detection while the rest rely on neutron detection. Due to attenuation, photon detection techniques are not as effective for assaying inner pins. Induced fission neutrons ~2 MeV are at high enough energy to pass through the fuel assembly and be counted at a detector. Therefore first initial importance maps were generated, using the MCNPX 2.7.A tally tagging feature, assuming neutron detection, in order to determine which fuel pins would be hardest to see in a diversion. Each NDA neutron detection system incorporates a series of detectors and structures that constitute an albedo at the detector fuel assembly interface that affects the multiplication in the fuel assembly. Furthermore the orientation of the detection equipment may further affect the importance of a given fuel pin in a given detection strategy.

For this study we developed an importance map, to assist in generating diversion scenarios, based on the Passive Neutron Albedo Reactivity (PNAR) counting system.[14] The system is composed of either He-3 tubes or fission chambers embedded in a block of polyethylene that surrounds the sides of the fuel assembly. The system also contains a cadmium sleeve that can be inserted in order to harden the spectrum of reflected leakage neutrons. PNAR assays total fissile content by examining the ratio of counting with and without the cadmium sleeve (consult Ref. 14 for details of the PNAR geometry). Therefore we examined the importance of particles with different albedos by investigating the difference in the importance maps under the different counting conditions. Though neutron emission rate is enrichment, burnup and cooling time dependent due to the buildup of ^{244}Cm and decay of ^{242}Cm, we chose to develop an importance map based on average fuel assembly characteristics. We developed importance maps based on a 3% enriched, 30 GWD/MTU burned, 1 year cooled assemblies. Therefore these counting conditions contain significant neutron contribution from ^{244}Cm spontaneous fission and slight contribution from ^{240}Pu and ^{242}Cm spontaneous fission.

Four types of counting conditions were examined: (1) PNAR counting in air with cadmium sleeve (ACd); (2) PNAR counting in 2000 ppm borated water with cadmium sleeve (BWCd); (3) PNAR counting in air without cadmium sleeve (ANoCd); (4) PNAR counting in 2000 ppm borated water without cadmium sleeve (BWNoCd). For each case the passive neutron source was generated assuming spontaneous fission neutron emission from the actinide content. Each case was run for 10 million histories such that the relative error of each tally tag did not exceed 3%. The tally tagging consisted of neutrons resulting from the following origins: (1) uncollided source neutrons that result detection; (2) source neutrons that scatter and result in detection; (3) induced fission neutrons that result in detection from both collided and scatter neutron flight paths. Fig. 2a displays the importance map for ANoCd, and Fig. 2b displays the importance map for BWNoCd. Fig. 2c displays the importance map for ACd, and Fig. 2d displays the importance map for BWCd. Each color within in each legend represents either 20%, 40%, 60%, 80%, and 100% of the range between least and greatest contribution.

The quantities displayed in the legend represent the unnormalized contributions to the total tally and therefore should be viewed in a relative sense.

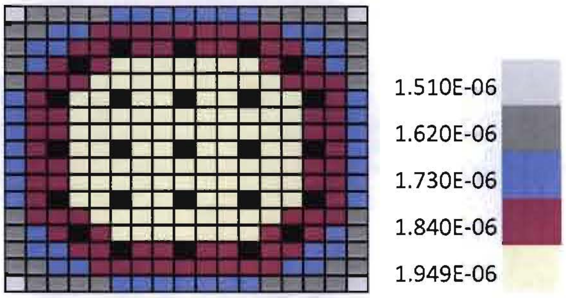

Fig. 2a. Importance map for ACd.

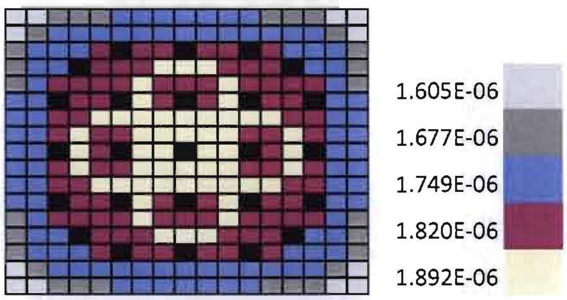

Fig. 2b. Importance map for BWCd.

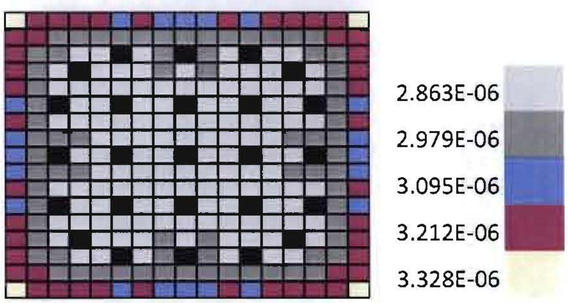

Fig. 2c. Importance map for ANoCd.

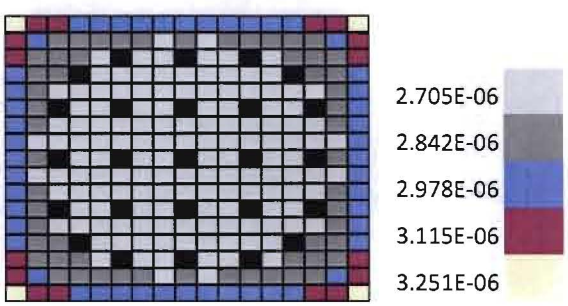

Fig. 2d. Importance map for BWNoCd.

The importance maps give insight into the physics incurred in the counting conditions. For cases ANoCd and BWNoCd, due to the excess moderation at the corners of the assembly, from having two adjacent pin sides facing the polyethylene, the corner pins posses the largest probability of being detected. The higher probability of detection is probably due to the larger burnup at the pin periphery and therefore larger higher actinide and ^{244}Cm emission as well as the increased moderation at the fuel assembly periphery leading to increased fission probability at this pin location. For cases ACd and BWcd, the outer pins are adjacent to a large absorber, and therefore less multiplication occurs in the outer pins because neutrons that are scattered out into the polyethylene and return thermal have a lower probability of returning due to thermal absorption in cadmium. Due to cadmium absorptions of emission neutrons, neutrons also have a lower probability of escaping the assembly region and entering the polyethylene for thermalizing. For example, comparing cadmium sleeve use versus no cadmium sleeve use, the percent difference in contribution of source neutrons that do not cause fission yet reach detector in air counting is ~26% while in water counting the percent difference is ~29%. The percentage difference in multiplication, comparing cadmium sleeve use versus no cadmium sleeve use, in air counting is 78% while it is only 63% in borated water.

Table I displays the range in importance of each fuel pin for each importance map. The range in importance for each air counting case, ACd and NoACd is similar; however, the range in importance for counting in BWNoCd is double that for counting in BWCd. This trend suggests that cadmium in the borated cases flattens out the importance in each fuel pin more significantly than the boron in air cases.

TABLE I

Range in Fuel Pin Contribution for Each Importance Map.

Case	Range	Difference
ACd	1.949E-06 – 1.400E-06	5.492E-07
BWCd	1.892E-06 – 1.533E-06	3.585E-07
ANoCd	3.328E-06 – 2.747E-06	5.811E-07
BWNoCd	3.251E-06 – 2.568E-06	6.829E-07

After considering results from the importance maps as well as considering the limitations of photon attenuation, several base diversion cases were generated. The decision was made to choose diversion maps consisting of diversions from 3 separate regions of the fuel assembly (inner region is the 7X7 inner square; middle region is outside the inner 7X7 square yet contained within the next set of guide tubes; the outer region is outside the final shell of guide tubes yet inside the most outside rows) . For each region 8, 24, and 40 pins were diverted represent 3.0%, 9.1%, and 15.2% of rods diverted. For each diversion case, initially LEU will be used as a diversion material due to the limiting attributes mentioned previously.

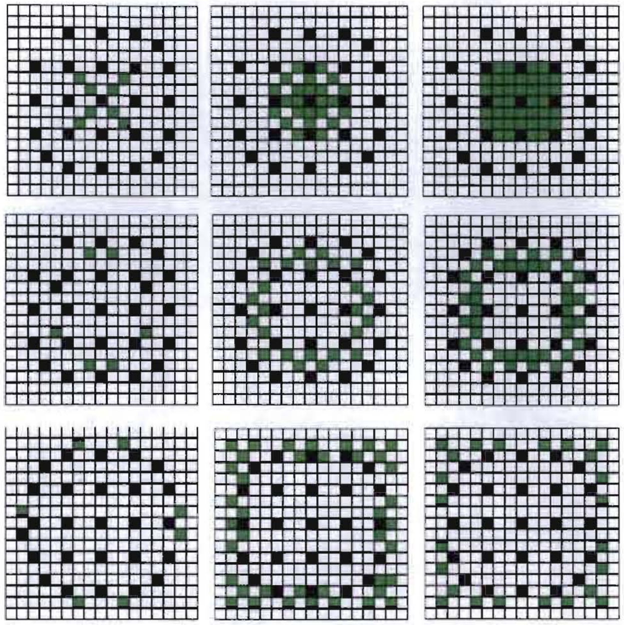

Fig. 3. Diversion Cases.

V. CONCLUSIONS

A large study is being undertaken to analyze the merits and faults of 12 different detection techniques in order to integrate a few techniques in order to determine the plutonium mass in spent fuel assemblies. Many facets of the study are completed, and tools have been generated in order to improve assessment efficiency. BAMF-DRT 1.0 was developed in order to automate the coupling of accessing MCNPX burnup out files, accruing information of importance, generating a fixed source assembly file for a given cooling time, wrapping a detector geometry around the assembly file, and generating diversion scenarios. BAMF-DRT uses the VBA platform within Microsoft Excel with visual user forms containing easy-to-use drop down menus and push button execution to minimize the time spent generating input files in order to maximize the time spent analyzing data. A spatial/enrichment/burnup /cooling time source term library has been generated using MCNPX 2.7.A for various initial enrichments, burnups, and cooling times expected to be encountered by the typical IAEA inspector. Diversion maps have been determined based on importance mapping using the tally tagging feature in MCNPX.

This paper describes the first step towards developing one of a few possible spent fuel NDA strategies. This study is also currently limited to generic PWR assemblies incorporating using MCNPX 2.7.A depletion. For the purpose of the down select process, the Westinghouse PWR 17 X 17 geometry is suitable to demonstrate base merits and faults of each technique. The base down-select will seek to determine an optimum integrated NDA strategy that allows the inspector the best possible method for assaying spent fuel. Peculiarities of each reactor type will be evaluated in the future. More assembly types, of various reactor designs, may be examined in future studies. Future efforts will require determining objective functions for comparing each NDA technique as well as building and analyzing hardware for each NDA technique.

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