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Monte Carlo Modeling of the Californium-Interrogation with Prompt Neutron (CIPN) Device for Spent Nuclear Fuel Measurements

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Abstract

Californium-Interrogation with Prompt Neutron detection (CIPN) is an active interrogation technique developed under the Next Generation Safeguards Initiative Spent Fuel (NGSI-SF) effort to improve capability to quantify the plutonium (Pu) mass in, and detect the diversion of pins from, spent nuclear fuel assemblies with non-destructive assay (NDA) techniques as well as estimate the initial enrichment, burnup and cooling time of the fuel. The CIPN detector consists of four neutron and two gamma-ray detectors in a compact, waterproof case. It is deployed on a pole and placed adjacent to an assembly in the spent fuel pool. A capsule of neutron-emitting californium-252 (Cf) is placed near the fuel assembly to induce fissions. The instrument is capable of measuring passive neutron background from the assembly as well as operating in the active-mode with the Cf source to analyze the fissile material content. Ion chambers can also provide a gross gamma count rate that can be related to the assembly burnup. Performance of the CIPN detector was evaluated with the Monte Carlo N-Particle transport code MCNPX using realistic spent fuel assembly libraries developed at Los Alamos National Laboratory (LANL). A large number of simulations provided data to correlate the detector response to specific assembly characteristics. These included the initial U-235 enrichment, irradiation history, and post-irradiation cooling time for each assembly. Future work will focus on integrating the CIPN detector signals to improve spent fuel assembly characterization and quantification of Pu mass.

Introduction

Spent fuel measurement technology plays an important role in nuclear safeguards. The application of safeguards is meant to provide “timely detection of diversion of significant quantities of nuclear material” and the “deterrence of such diversion by the risk of early detection”. [1] The National Nuclear Security Administration (NNSA) supports the development of safeguards technology through the Next Generation Safeguards Initiative (NGSI). The NNSA sponsors NGSI as “a comprehensive initiative to revitalize the international safeguards technology and human resource base.” [2]

The CIPN instrument is being developed within the NGSI Spent Fuel (NGSI-SF) effort for the purpose of measuring spent fuel. NGSI-SF is a multi-laboratory, multi-university project that supports the development of novel detector systems to improve spent fuel safeguards. Its primary goals are to develop Non-Destructive Assay (NDA) instruments that can collectively perform several key functions. Most important is the ability to detect missing or replaced pins from spent fuel assemblies (SFAs) to confirm item integrity and deter diversion. Next is the capability to determine plutonium mass and related plutonium and fissile mass parameters in SFAs. It is also essential to verify the facility-declared values of initial enrichment (IE), burnup (BU), and cooling times (CT) for SFAs. [3] In this nominally five year effort, 14 possible instrument systems were evaluated for development based on effectiveness, cost, ease of use, and other merits. [4] This vetting process has removed several techniques from development. Along with CIPN, current techniques include Differential Die-Away (DDA), Differential Die-away through Self-Interrogation (DDSI), Passive Gamma (PG), Total Neutron (TN), Passive Neutron Albedo Reactivity (PNAR), and Self-Interrogation Neutron Resonance Densitometry (SINRD).

This paper focuses on detailed MCNP evaluations of CIPN performance with simulated spent fuel assemblies. Detector response was assessed using a broad range of assembly characteristics, with burnups of 15-60 GWd/MTU, initial enrichments of 2-5%, and cooling times up to 80 years. This work is a follow-up to previous MCNP evaluations of the initial CIPN design with an earlier generation of simulated spent fuel assemblies. [5] The key contribution of this work is the extensive performance evaluation of the final CIPN instrument design. These MCNP simulations examined 66 spent fuel assemblies and covered a much broader range of characteristics than measurements could. Following MCNP characterization, the initial CIPN design was modified for experimental testing in the Republic of Korea (ROK). The results from this field trial will be published separately. For the purpose of uncertainty quantification, this work uses experimentally-evaluated systematic errors.

The CIPN Instrument

CIPN comprises an integrated instrument capable of measuring the gamma and neutron radiation emitted by spent fuel assemblies. Four neutron and two gamma-ray detectors measure the neutron count rate and gamma-ray energy deposition rates, respectively. A U-shaped high density polyethylene (HDPE) case provides waterproofing for the electronics and a moderator for the neutron detectors. Signal cables pass through the back of the instrument and connect to monitoring equipment outside of the pool. **Figure 1** diagrams the device next to a fuel assembly.

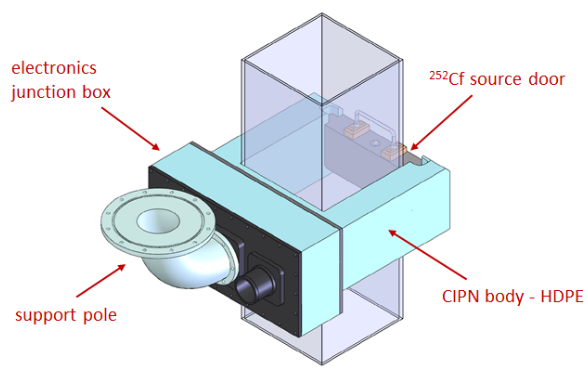

Figure 1: Diagram of the CIPN Instrument

Both passive and active measurements are performed with the CIPN instrument to yield three independent signals. The gross Passive Gamma (PG) and Total Neutron (TN) signals are measured simultaneously. The TN signal provides a baseline neutron count for the SFA being assessed. An active measurement with ^{252}Cf interrogation measures the CIPN signature from induced fissions plus the TN signal. Neutrons are measured with fission chambers (FCs) and gamma-rays are measured with ion chambers (ICs). The original CIPN instrument did not include the ability to measure PG. [5] [6] The two ion chambers were added later to provide additional measurement capabilities without significantly increasing the instrument complexity.

CIPN Signature

The CIPN signature is produced by placing a neutron source next to the assembly of interest. Fission is induced in the assembly and neutron multiplication occurs. Some of the neutrons produced by fission are leaked from the assembly and detected in fission chambers (FCs). Since the TN signature is very large in the assembly, the CIPN signature must be strong relative to this passive signal. **Figure 2** illustrates the passive TN and net active CIPN measurements.

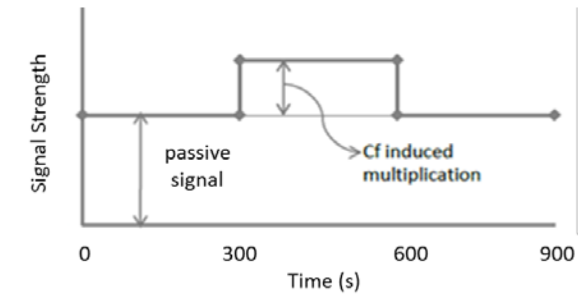

Figure 2: Conceptual Diagram Showing Passive and Active Interrogation Signals

As shown in in **Figure 2**, the CIPN signature is superimposed on top of the total neutron signal. The net signal is calculated by subtracting the TN count rate from the active count rate. To distinguish from the instrument itself, the CIPN signature is referred to in this paper as the net active signal.

The instrument achieves a very high count rate due to the optimized detector layout and ^{252}Cf source strength. It also has a relatively uniform detector response from each region of the fuel. CIPN performance was evaluated using fresh fuel at Los Alamos National Laboratory (LANL) as well as with four spent fuel assemblies in the Republic of Korea (ROK). [7] Field trials demonstrated that CIPN can achieve counting uncertainties of less than 1% in a few minutes of measurement time for the range of assemblies measured. These included three assemblies with BUs of 28, 36, and 38 GWd/MTU with 3.2% IE and about 25 years of CT, plus one assembly with 17 GWd/MTU BU, 2.1% IE and a 34 year CT. [8]

MCNP Modeling

The CIPN instrument was evaluated in MCNP using simulated spent fuel libraries (SFLs) representing isotopic pin compositions of 17x17 pressurized water reactor (PWR) assemblies. These were produced specifically for the NGSI-SF effort. The libraries contain sets of fuel assemblies with varying initial enrichment (IE), burnup (BU), and cooling times (CT). A wide range of assemblies was used to characterize detector performance over many possible scenarios.

The initial instrument design was evaluated using Spent Fuel Library 1 (SFL1). [5] [6] This library contains PWR assemblies with symmetric burnup profiles across the assembly. The fuel profile is axially homogenous and includes about 80 isotopes within the fuel. [9] Despite its simplicity, SFL1 gave good representations of the CIPN signal response for initial performance evaluations. Once the CIPN instrument design was finalized and PG capabilities were included, it was modeled using a subset of Spent Fuel Library 2 (SFL2). This subset, called SFL2a, has asymmetric burnup profiles with a fixed fuel shuffling scheme. SFL2a contains axially homogenous assemblies with approximately 130 isotopes per fuel pin. [9] Assemblies covered a range of characteristics, with BU values of 15, 30, 45, or 60 GWd/MTU, IE values of 2, 3, 4, or 5%, and CT values of 14 days, or 1, 5, 20, 40 or 80 years.

Simulations were conducted on MCNPX 2.7.4, a LANL-only version of MCNP. This version contains several features that were developed for safeguards instrumentation research. One such feature is the First Fission tally (FFT), which is useful in active interrogation techniques. This tally modifies an existing MCNPX tally by identifying the first nuclide to fission in a fission chain. [10] FFT was used here to assess the direct contribution of ^{252}Cf source neutrons to the net active CIPN signature. This behavior is important to quantify but can be difficult to measure experimentally.

Figure 3 shows an MCNP model of the CIPN instrument placed around a 17x17 PWR assembly. The electronics housing and frame mount are omitted from the simulated design.

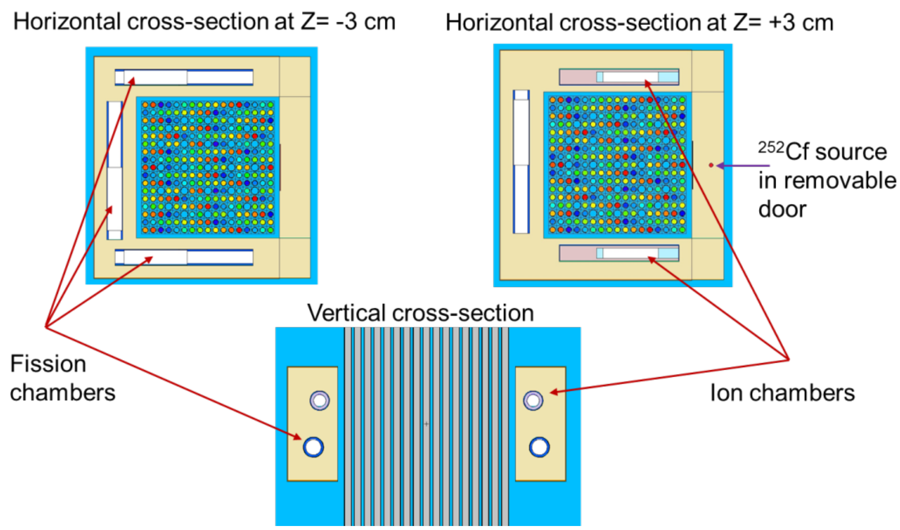

Figure 3: Cross Sections of the CIPN Instrument with ^{252}Cf Source in Position

The fuel pins in **Figure 3** are shaded to distinguish the different material types in MCNP. Electronics and support structures are omitted from the MCNP model.

The net active CIPN response was modeled using a point source of isotropically emitted neutrons. Neutron energies were sampled from the Watt's fission spectrum for ^{252}Cf spontaneous fission emissions. Weight windows were used to reduce computation time and improve convergence. The F4 tally monitored the cell-averaged flux in the fission chambers. F4 tally results were converted directly into counts per ^{252}Cf source particle by using segment divisor (SD) and tally multiplier (FM) cards. These accounted for the volume and macroscopic fission cross section of the uranium coating in the fission chambers.

Passive gamma and total neutron simulations were performed separately from the net active interrogation simulations. TN signals were evaluated using the same tallies as the net active signal, while gamma signals used the F6 tally. This measured the cell-averaged energy deposition from incident gammas in the ion chambers. Results were converted to energy deposition rate using the SD card to account for the mass of fill gas in the detector. F4 tallies were applied to the ICs to verify that the F6 tallies were functioning as desired. Both TN and PG used weight windows for variance reduction, and PG also used source biasing. Both passive simulations used source terms that were volumetric and homogenously distributed within the fuel pins. Cladding activation was ignored. All assemblies were modeled for the TN response, and a representative sample of 17 assemblies was modeled for the PG response. Through previous work in NGSI-SF, the PG emissions from all SFL2a assemblies had been characterized in MCNP for xenon-filled ion chambers. [11] It was found that the PG response in the CIPN instrument differs from the previous PG work by a factor of 4.97 ± 0.02. Since the CIPN instrument PG response is linearly scaled to the previous results, the PG data presented here is drawn from the earlier PG data and is scaled by a factor of 4.97.

Count rates are obtained from the modified MCNP tallies by multiplying tally outputs by source strengths values. The net active CIPN signature is the product of the total fission chamber response and the ^{252}Cf source strength. Since a 60 µg source was used in the ROK measurements, this same source strength is used here although past research has shown that a source 2 to 3 times stronger will be needed for typical commercial fuel. [5] Count rates are obtained for TN and PG signals by multiplying the MCNP results by the total number of neutrons or photons emitted per second, respectively.

Uncertainty Propagation

Error was propagated using four different uncertainties. MCNP uncertainties quantify the degree of convergence in the MCNP simulations. Counting uncertainty comes from the Poisson behavior of radioactive decay, and decreases as the number of counts increase. Rotational uncertainty occurs because signal strengths vary depending on which side of the assembly is measured. This occurs in SFAs with horizontally asymmetric BU gradients. Positioning uncertainty results from variations in the spacing between the assembly and the arms of the CIPN instrument. Experimental evaluations have found that rotational uncertainty can vary from mean values by 5% or less in TN and by less than 2% in PG and net active CIPN. It was found that the net active signature is relatively invariant even with horizontal burnup gradients of 9% between quadrants. Estimates of positioning uncertainty have been made for gap variations of 1 cm between the assembly and CIPN walls. TN positioning uncertainties are below 5%. PG and net active positioning uncertainties fall below 3%. Once all four uncertainties have been quantified, the total expected measurement error is calculated using the square root of the sum of the variances.

MCNP Results

MCNP results are shown in three plots that illustrate the sensitivity of detector signals to SFA parameters. Statistical uncertainties of the two neutron signals are representative of 300 second long measurements using a 60 µg ^{252}Cf source. The passive gamma signal is displayed as a rate, with electrical current on the Y-axis.

Figure 4 shows the total expected signal in a five minute measurement of 30 GWd/MTU SFAs. **Figure 5** shows measurements of 4% IE SFAs. Some error bars are smaller than the size of symbols.

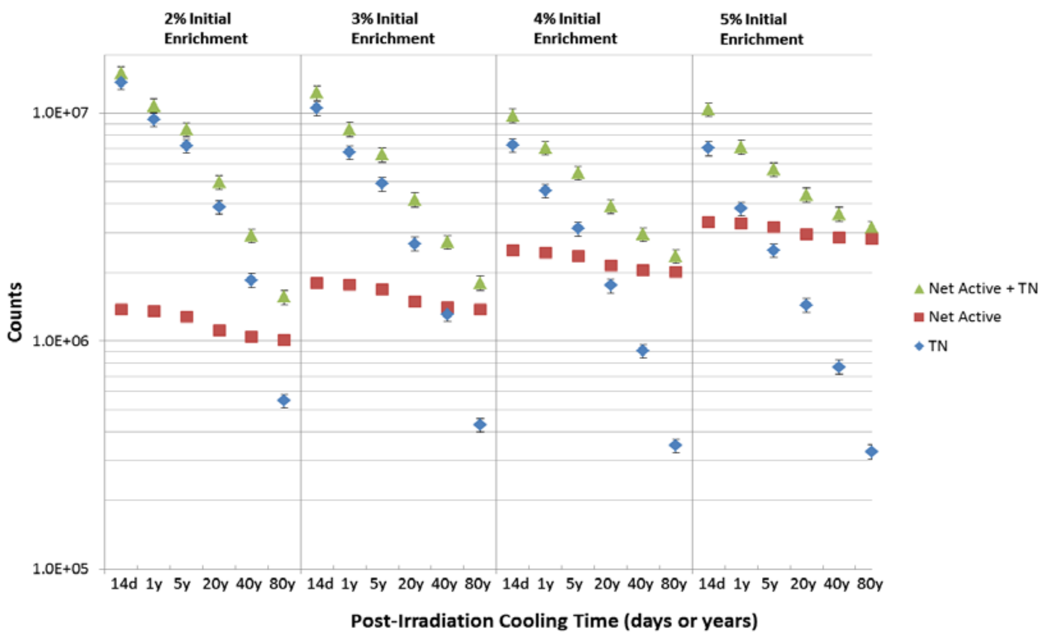

Figure 4: Expected Neutron Counts in a 300 Second Measurement of 30 GWd/MTU Assemblies

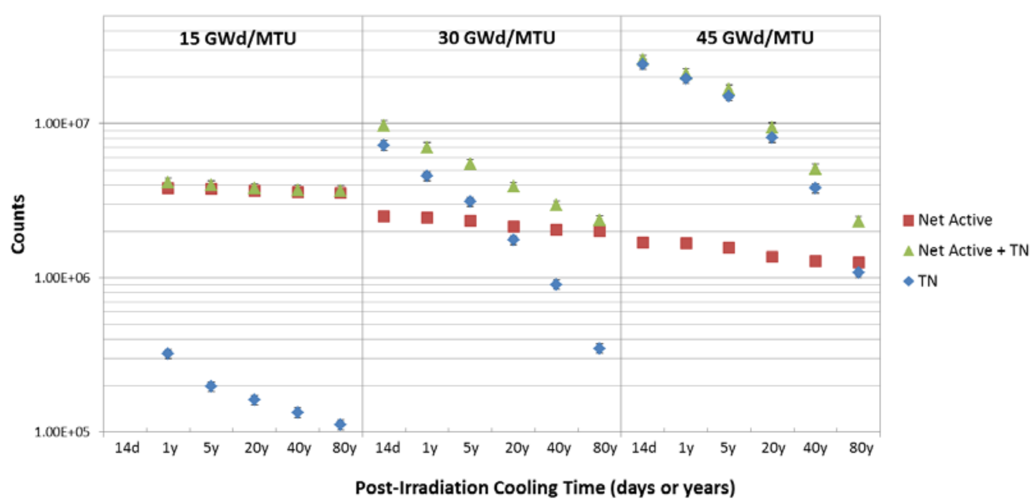

Figure 5: Expected Neutron Counts in a 300 Second Measurement of SFAs with 4% Initial Enrichment

Figure 4 and **Figure 5** illustrate the sensitivity of the TN and net active CIPN signature to various assembly characteristics. The net active signal has a strong dependence on IE, BU, and CT. All parameters influence TN, but changes in CT exhibit the greatest effects on the TN signal.

Figure 6 shows the expected current in microamperes (μA) for measurements of fully-burned assemblies. Error bars are about 3% and are smaller than the size of symbols.

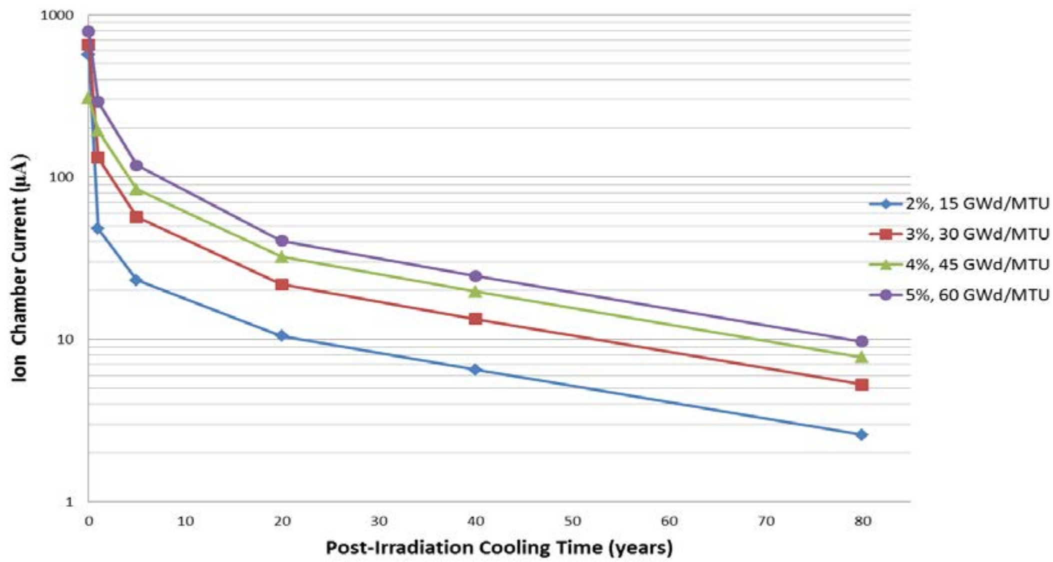

Figure 6: Expected Current in Ion Chambers from Fully-Burned Assemblies

Figure 6 shows the strong dependence of the PG signal on CT and BU. Differences between the four curves are largely due to BU and not IE. For CT values greater than 5 years, IE has little effect on the PG signal.

The First Fission Tally (FFT) in MCNP was used to determine what portion of the net active signature came directly from ^{252}Cf source neutrons. **Table 1** shows how signal contributions vary with IE and BU in 5 year CT assemblies.

Table 1: Contribution of ^{252}Cf source neutrons to the net active CIPN signature for select 5 year CT SFAs

	Percent of Contribution	Error	Rate of contribution (s^{-1})	Error
2% IE, 15 GWd/MTU	11.14%	0.03%	6.90E+02	0.09%
3% IE, 30 GWd/MTU	12.11%	0.03%	6.77E+02	0.09%
4% IE, 15 GWd/MTU	5.25%	0.01%	6.63E+02	0.08%
4% IE, 30 GWd/MTU	8.43%	0.02%	6.61E+02	0.09%
4% IE, 45 GWd/MTU	12.79%	0.03%	6.68E+02	0.09%
5% IE, 60 GWd/MTU	12.75%	0.03%	6.61E+02	0.10%

In all assemblies, some of the net active signal results directly from neutrons emitted by the ^{252}Cf source. Although the relative streaming contribution can reach up to 21% for some assemblies, the total contribution remains fairly constant around 670 counts per second.

Discussion

Several trends in the data can be observed. As the time after irradiation increases, radionuclides decay and daughter products accumulate. Increased CT results in reduced signals for PG, TN, and net active signature. This occurs as radionuclides like ^{154}Eu, ^{134}Cs, ^{137}Cs, ^{242}Cm, ^{244}Cm, and ^{241}Pu decay. Changes that result from increased BU differ between the active and passive signals. Both passive signals increase with BU while the net active signature tends to decrease. The PG signal is directly related to the buildup of fission products in the fuel. Similarly, the TN signal results from the buildup of actinides with high spontaneous fission decay probabilities. The net active signature changes as ^{235}U is consumed and Pu and neutron absorbing isotopes accumulate. ^{235}U contributes the most to the signal at low BU, but this wanes as enrichment decreases throughout the life of the assembly. The concentration of ^{239}Pu increases over the first few tens of GWd/MTU and then flattens out. ^{241}Pu increases throughout the burn cycle. The net result is that the net active signature decreases with increasing BU. Signal responses from changes in IE tend to vary. PG signals remain relatively static when IE changes. This is largely due to the identical ^{137}Cs fission yields from ^{239}Pu and ^{235}U. The net active signature becomes larger with higher IE because more ^{235}U can contribute to the signal. Conversely, TN decreases as IE increases. This occurs with changes in the macroscopic absorption and fission cross sections, Σ_a and Σ_f. When IE increases, Σ_f becomes larger without significantly affecting Σ_a. Since power is kept constant during the assembly irradiation histories, neutron flux must decrease when Σ_f increases. The smaller flux results in a lower ratio of absorption to fission interactions as IE increases. [12]

Each assembly produces a unique combination of PG, TN, and net active CIPN signals that allows it to be distinguished. Previous researchers including J. Hu, V. Henzl, A. Favalli, et al. have developed semi-empirical relations using SFL1 results to solve for IE, BU, CT, as well as elemental plutonium content. [5] [6] [13] [14] An additional approach by Tom Burr et al. has used exploratory data analysis to assess relations between assembly parameters and measured signals. [15] Results from using SFL2a will be used to train a numerical solver. The performance of this solver will be assessed using results from Spent Fuel Library 4 (SFL4). In order to preserve the integrity of the performance testing, the characteristics of SFL4 assemblies are not revealed to the modelers. This is done with the intention of preventing human biasing in the data fitting.

CIPN also has some capability to assist in detecting pin diversions. Assessments by Jianwei Hu et al. have determined that when baseline measurements have been made, a diversion of as few as eight pins can be detected. This assumes that spent fuel pins are replaced with dummy pins containing depleted uranium, and that measurement uncertainties are approximately 1%. [5] [6] However, experiments have shown that actual measurement errors are larger than this. It is likely that as many as 24 pins must be removed before CIPN can detect a diversion.

Conclusions and Future Work

The CIPN instrument offers integrated capabilities for applying safeguards to spent nuclear fuel. It measures three signals that are sensitive to the initial enrichment (IE), burnup (BU), and cooling time

(CT) of a spent fuel assembly (SFA). Each SFA yields a unique combination of Total Neutron, Passive Gamma, and net active californium interrogation signals. Researchers have previously shown that CIPN can detect partial diversions of spent fuel assembly pins; however, pre-diversion baseline measurements are necessary. Exploratory data analysis will be used to quantify the effects of IE, BU, and CT on the three measured signals. Quantification and propagation of uncertainty will be used together with model validation to give confidence in data analysis results. Correlations between the three SFA parameters and the total SFA plutonium mass will be examined, as well as methods to detect partial diversions without pre-diversion baseline measurements.

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