LA-UR- 11-05673

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Intended for: Conference of Global 2011
Makuhari, Japan
Dec. 11-15, 2011

Los Alamos NATIONAL LABORATORY EST. 1943

Quantifying Fissile Content in Spent Nuclear Fuel Assemblies Using ^{252}Cf Interrogation with Prompt Neutron Detection

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Abstract

The majority of the plutonium on earth is stored in spent nuclear fuel assemblies. Presently, there is no means for directly measuring the mass of the plutonium in these assemblies by nondestructive assay (NDA). Researchers at LANL have been coordinating a multi-laboratory effort to quantify the capability of 14 NDA techniques for the purpose of combining a subset of these techniques into a system that can directly measure the isotopic Pu mass. ^{252}Cf Interrogation with Prompt Neutron (CIPN) detection is one of the 14 proposed NDA techniques, and it shows promise of quantifying fissile content in spent fuel assemblies. CIPN is a relatively low-cost and portable instrument, and it looks like a modified fork detector combined with an active interrogation source. Fission chambers were chosen as the neutron detectors because of their insensitivity to γ radiation. The CIPN assay comprises two measurements, a background count and an active count, without and with the ^{252}Cf source next to the fuel respectively. The net signal above background is primarily due to the multiplication of Cf source neutrons caused by the fissile content. It is almost uniformly sensitive to diversions at different locations aross the assembly. A 100-μg ^{252}Cf source was proven strong enough to provide sufficiently high signal above background. The concept of $^{239}\mathrm{Pu}_{\mathrm{eff\_CIPN}}$ was introduced to represent the three major fissile isotopes in a single term. Burnup (BU) and cooling time (CT) corrections were introduced to $^{239}\mathrm{Pu}_{\mathrm{eff\_CIPN}}$ to account for the neutron absorption caused by different neutron absorbers. The results shows that there exists a coherent universal relation between CIPN count rate and "adjusted fissile content". With the schemes presented in this paper, together with given BU and CT (or quantified using other techniques), the fissile content of the target spent FA (or $^{239}\mathrm{Pu}_{\mathrm{eff\_CIPN}}$) can be determined within a few percent. At the end, the conceptual experiment setup of CIPN is also briefly discussed.

Keywords: fissile content, spent fuel, plutonium, nuclear safeguards, CIPN, NGSI, MCNP, NDA

1. Introduction

Non-destructive assay (NDA) techniques have been applied to characterize spent nuclear fuel for several decades. In 2009, the Next Generation Safeguards Initiative (NGSI) of the U.S. Department of Energy's National Nuclear Security Administration (NNSA) began a five-year effort to develop an integrated instrument fully capable of determining Pu mass in, and detecting diversion of pins from, commercial nuclear spent fuel assemblies (SFA) [1, 2]. Fourteen NDA techniques were proposed and investigated, and ^{252}Cf Interrogation with Prompt Neutron detection (CIPN)is one of them. None of these techniques, in isolation, is capable of quantifying the Pu mass in an SFA, so it requires the integration of several techniques to piece together the information provided by each individual technique.

CIPN shows promising capability of quantifying fissile content in an SFA. CIPN is also a portable and inexpensive (<\$350k) detector. CIPN is similar to a Fork detector, which has been used in the field for decades to measure total neutron and gamma emission, in both size and shape [3]. Due to the limitations of the Fork detector, an active interrogation source (^{252}Cf) is introduced next to the SFA on the opposite side to the detector, which forms the basic concept of CIPN. ^{252}Cf source has been widely used in NDA instruments because of its high neutron intensity, portability and reliability. It has the intensity of a small accelerator without the electronics and irregular variations in yield. One particular example is a shuffler, which measures delayed neutrons using a 500-μg ^{252}Cf source [4]. Fission chambers (FCs) are chosen as neutron detectors because of their insensitivity to gamma radiation. The geometry and physics of CIPN were modeled by using MCNPX 2.6.0 [5]. The CIPN detector has been simulated with the NGSI spent fuel library [6] to quantify its capabilities of determining fissile content and detecting diversion of fuel pins in an SFA. The library covers a wide range of parameters, i.e., burnup (BU), initial enrichment (IE), and cooling time (CT), found in PWR SFAs. From the library, 64 SFAs were selected, and each of them represents a unique combination of BU (15, 30, 45 and 60 GWd/tU), IE (2, 3, 4, 5%) and CT (1, 5, 20 and 80 years).

2. The Concept of CIPN Assay

In Fig. 1 a conceptual diagram of CIPN signal composition is shown. The CIPN assay is comprised of two measurements, a background count and an active count. During

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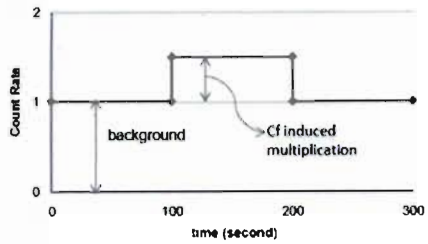
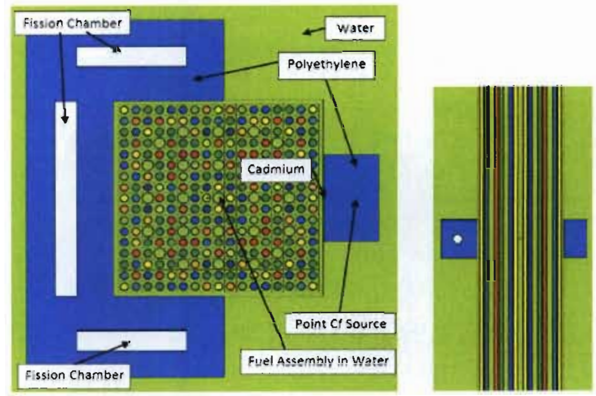


Fig. 1: Diagram of conceptual CIPN signal composition (background and active assay).



(a) (b)

Fig. 2: (a) Horizontal cross section of CIPN; (b) Vertical cross section of CIPN.

an active count, the ^{252}Cf source is moved next to the assembly where it remains stationary for $\sim$100 seconds. The background count is from two sources: 1) direct contribution of passive source neutrons, namely spontaneous fission neutrons (mainly from ^{244}Cm and ^{242}Cm) and (α, n) reactions with ^{18}O etc. [7]; 2) the multiplication of passive source neutrons due to fissile content in an SFA. In addition to background sources, two more sources contribute to the active count: 1) direct contribution of the ^{252}Cf source neutrons; 2) the multiplication of ^{252}Cf source neutrons due to fissile content in an SFA. With optimized design of the detector, the contribution of direct Cf neutrons counts only a small fraction of the signal in water (or borated water). Hence, the increase in the count rate above background count when the ^{252}Cf source is introduced is almost entirely from multiplication of the fuel. To relate multiplication to fissile content, corrections are needed to account for absorptions caused by neutron absorbers. Besides, the increase in signal brought by the active source has to be large enough to override the background signal to reduce statistical uncertainty, meaning the ^{252}Cf source is required to have sufficient strength.

3. Design of CIPN Detector

Besides to make CIPN a portable and low cost detector, one additional goal is to achieve uniform sensitivity across the whole fuel assembly (i.e., if same amount of fuel is diverted from different locations within an SFA, the detector is expected to respond similarly). To achieve these goals, a few design variables were optimized: 1) the Cf source location was optimized to (a) ensure the source neutrons are appropriately moderated before entering the fuel assembly, which reduces ^{238}U fission and emphasizes fissile isotopes, and to (b) ensure that the solid angle is large enough for the source neutrons enter all along the side of the SFA; 2) the arrangement, length and position of FCs were optimized to ensure uniform sensitivity across the whole SFA; 3) the thickness of polyethylene around FCs was optimized to maximize the count rate.

After comparing several different designs and running $\sim$200 MCNPX simulations, the design of CIPN was finalized with aforementioned optimized variables. Fig. 2(a) shows the horizontal cross section of CIPN, with a 17×17 PWR SFA located at the center (the vertical view is shown in Fig. 2(b)). The medium surrounding the pins within the assembly may be water, borated water or air. (Water was chosen for the purpose of this study.) Surrounding the assembly on all sides is a 0.5-*cm* gap filled with water, to provide the mechanical tolerance for moving an assembly through the detector. The blue rectangular to the right most is a block of polyethylene that contains a single-point ^{252}Cf source (which releases intense neutrons with average energy of $\sim$2.3 *MeV* from spontaneous fission). The distance from the source to the edge of the assembly is $\sim$4 *cm*. There is a thin cadmium sheet (1-*mm* thin, not visible in the figure) inserted between the assembly and the polyethylene block to even out the fission rate in the assembly along the vertical direction. The vertical "U"-shaped blue block is filled with polyethylene embedded with 3 fission chambers, which have 93%-enriched ^{235}U ultra thin layers (1-μm thin) coated inside the 1"-diameter aluminium tube. The thickness of the polyethylene is $\sim$3 *cm*. The polyethylene block has a cadmium liner all around except the walls directly facing the assembly or the source, in order to minimize signal from background neutrons.

Variance reduction techniques (e.g., weight window) were used in MCNPX simulations to improve calculation efficiency by reducing the sampling of neutrons that are far away from the detector, since the assembly is 3.8-meters long and the detector occupies only 10 *cm* of its mid section. Fig. 3 shows the count rate changes in percentage against the base case (no rods diverted) when 12 rods diverted at 6 different zones in an assembly each time (for this particular exercise, all the fuel rods are filled with fresh UO_2 (2% enrichment)). In each of the 6 cases, 12 rods (either all fuel rods or 11 fuel rods plus 1 water tube in some cases), which account for $\sim$4.5% of total fuel, in one of the 6 zones were replaced by fuel rods filled with depleted uranium (with 0.2% ^{235}U). Then the count rate was compared to the base case. As depicted in this figure, CIPN is sensitive to diversion because given the MCNPX uncertainty is less than 0.5% (the actual counting statistic is better than this) and lowest signal change is 7.3%, the minimum detectable diversion is well below 12 fuel rods. As shown in Fig. 2(a), the FCs are arranged in such a way to achieve uniform sensitivity. If the FCs are only on the left side, the count rate change of the top 3 zones (in Fig. 3) will be drastically reduced. The FCs on the top and bottom are shortened and

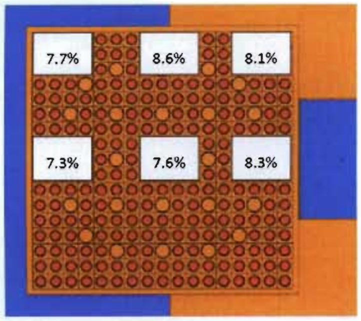


Fig. 3: Count rate changes against the base case when 12 rods diverted at 6 different zones in a FA.

shifted 9.3 *cm* to the left from the center, so the area closer to the source in the assembly will not be overly-emphasized.

4. CIPN Background Signal and Source Strength Requirement

As we know, in absence of an active source, an SFA gives out neutrons caused primarily by spontaneous fission of transuranic isotopes, such as ^{244}Cm and ^{242}Cm. These isotopes accumulate during the burnup of the fuel. A complete study of the background neutron source strength for all 64 SFAs were done by Richard et al. [7], and it also includes (α, n) neutrons. Given same background source strength, two different SFAs will have different background count rates in CIPN due to different multiplications. In order to quantify the Cf source strength requirement, the background count rate has to be quantified.

Four SFAs were chosen for this study: they have the same IE and CT, 4% and 5 years respectively, but four different BU (15, 30, 45, and 60 GWd/tU respectively). Since the background mainly scales with BU, despite of slight dependence on IE and CT as well, these 4 SFAs essentially cover the full range of background variation for the entire library. For the background counting (*B*), the $par = SF$ function in MCNPX was used to enable the sampling of source neutrons from spontaneous fission of transuranics. In this calculation, the spontaneous-fissioning isotopes are assumed distributed uniformly within the fuel assembly. Later study shows that this simplification would cause ~1% error in count rate comparing to a more realistic sampling (when the atom density of ^{244}Cm varies on both inter- and intra-pin level in accordance to burnup results). Net count rates (*S*) for these 4 SFAs were also obtained using MCNPX simulations.

Table 1 shows the background count rate (CR) and active count rate of the 4 SFAs. A 100-μg ^{252}Cf source was used for the active assay and it releases 2.34E8 neutrons per second. As shown in this table, on "per source neutron" base, the count rate (or "CR/n") in both background and net signal decreases with BU. This is due to two main factors: (1) less fissile materials and (2) more neutron absorbers in SFAs with higher BU. The background source strength increases dramatically with BU, as a results, the total background count rate increases with BU. While for net signal, the source strength

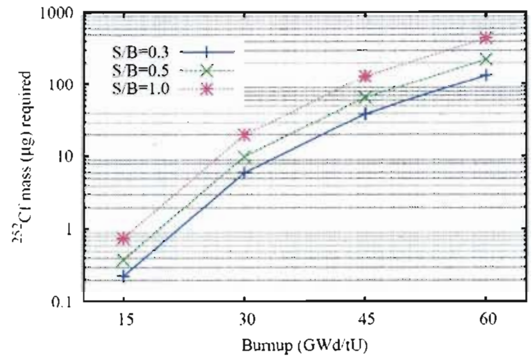


Fig. 4: Mass of ^{252}Cf (μg) required to achieve certain Signal-to-Background (S/B) ratios as a function of burnup.

remains the same for all 4 cases, and thus the total net count rate decreases with BU mainly due to reduced multiplication in SFAs with higher BU. This table also shows that the 100-μg ^{252}Cf source is sufficient since the count rate will increase at least 22% with the presence of the Cf source, given the counting uncertainty is less then 0.3% [8]. Fig. 4 shows the required mass of ^{252}Cf (μg) to achieve certain Signal-to-Background (S/B) ratios as a function of burnup. In general, larger source is needed for higher BU. It was decided to use the BU of 45 GWd/tU as a reference case for source selection since it is a higher BU than the vast majority of SFAs experienced by inspectors in the field. As illustrated in this figure, a 40-μg ^{252}Cf source will produce a signal of 30% above the background, and a 60-μg ^{252}Cf source will produce a signal of 50% above the background for 45 GWd/tU. Given these two bounding cases, a 50-μg ^{252}Cf source was determined as needed at the end of life for the source. Tt is desirable for the source to last at least five years, given the 2.65 year half-life of ^{252}Cf, a 200-μg Cf source was selected as the mass at the beginning of operation. Unless stated otherwise, the mid-life mass of a 100 μg (2.34E8 neutrons per second) was selected for results quoted in the remainder of this paper. As a point of reference, a 55-gallon-drum shuffler usually begins operation with a 550-μg source [4]. Furthermore, the largest ^{252}Cf source that are commercially available is 10,000-μg [9].

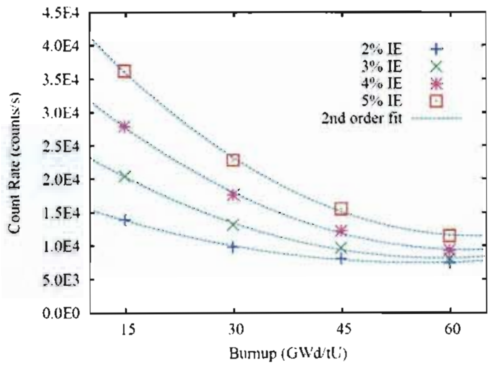
5. Preliminary Analysis of CIPN Signal

Before a more involved analysis is performed, it is instructive to illustrate how the count rate (CR) changes with the three main variables that make each of the 64 SFAs unique: BU, IE, and CT. Given the wide variation of these parameters, it is helpful to freeze one variable while looking at the change of count rate with other variables. In this section, two cases have been studied:

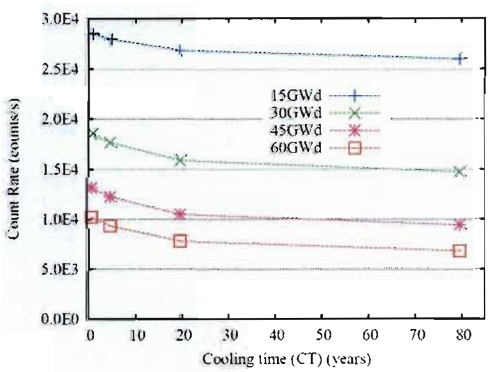
- Case 1, CR vs. BU, and the CT is held constant at 5 years. The count rates of 16 different assemblies (they are all 5 year cooled but with different BU or IE) are quantified as a function of BU and CT.
- Case 2, CR vs. CT, and the IE is held constant at 4%. The count rates of 16 different assemblies (they all have

BU	Background (B)			Net Signal (S)			S/B
(GWd/tU)	CR/n	source (n/s)	total CR	CR/n	source (n/s)	total CR	–
15	9.69E-5	2.95E6	2.85E2	1.61E-4	2.34E8	3.77E4	132
30	6.86E-5	7.03E7	4.82E3	1.03E-4	2.34E8	2.41E4	5.01
45	5.41E-5	4.10E8	2.22E4	7.25E-5	2.34E8	1.70E4	.77
60	4.70E-5	1.24E9	5.83E4	5.57E-5	2.34E8	1.30E4	.22

Table 1: The background count rate (B), net count rate (S) and the ratio (S/B) of the 4 SFAs with 4 different BU, and the same IE (4%) and CT (5 years). Note "CR" stands for count rate (counts/second), "CR/n" stands for count rate per source neutron and "source (n/s)" stands for neutron source strength in an SFA. A 100-μg ^{252}Cf neutron source was used in active assay with an intensity of 2.34E8 n/s. The background source strength was calculated by Richard et al. [7].



(a) Case 1



(b) Case 2

Fig. 5: (a) Case 1: the CIPN net count rate as a function of BU for 16 different assemblies in water (with CT fixed at 5 years); (b) Case 2: the CIPN net count rate as a function of CT for 16 different assemblies in water (with IE fixed at 4%).

4% IE but different BU or CT) are quantified as a function of BU and CT.

As shown in Fig. 5(a), the CIPN net count rate varies significantly with both BU and IE (Case 1). Following a particular curve of the same IE, the count rate decreases with BU because SFA with higher BU has lower fissile content and more neutron absorbers. For a given BU, SFA with higher IE has higher count rate because it has higher fissile content. The impact of different IE on count rate decreases at higher BU.

In Fig. 5(b), the variation in the CIPN net count rate with CT is illustrated (Case 2); the IE was held constant at 4%. As shown, the count rate decreases with longer CT but to a lesser degree than the variation with BU and IE. The observed change is due to two primary factors: (1) the fissile isotope ^{241}Pu, with a half-life of $\sim$14 years, decays into a pure neutron absorbing isotope ^{241}Am (with a significant neutron absorption cross-section); (2) the stable isotope ^{155}Gd, which has a "huge" neutron absorption cross-section (e.g., 10,000 barns at 0.1 eV) grows into assemblies since it is a decay product of ^{155}Eu (half-life of 4.68 years). The count rate drops at a slower pace at high CT because there are less ^{241}Pu and ^{155}Eu remaining to decay at higher CT. Hence, both ^{241}Am and ^{155}Gd accumulate over time, depressing the count rate at higher CT. The four curves in this figure are almost parallel to each other because the time dependence of the relevant factors are similar.

6. Further Analysis of CIPN Signal with $^{239}\mathrm{Pu}_{\mathrm{eff}}$

6.1. *The concept of* $^{239}\mathrm{Pu}_{eff}$

The fissile content is the main factor affects the count rate. In an SFA, there are three major fissile isotopes: ^{235}U, ^{239}Pu and ^{241}Pu. Inspired by the convention in the safeguards profession of using the concept of $^{240}\mathrm{Pu}_{\mathrm{effective}}$ for passive Pu coincident neutron counting [3], a similar term, $^{239}\mathrm{Pu}_{\mathrm{eff\_CIPN}}$ was introduced to represent weighted lump-sum of the 3 fissile isotopes. $^{239}\mathrm{Pu}_{\mathrm{eff\_CIPN}}$ is defined here:

$$^{239}\mathrm{Pu}_{\mathrm{eff\_CIPN}} \equiv C_1{}^{235}\mathrm{U}_m + {}^{239}\mathrm{Pu}_m + C_2{}^{241}\mathrm{Pu}_m. \quad (1)$$

In words, Eq. 1 weights the mass of ^{235}U and ^{241}Pu by constants C_1 and C_2, respectively, relative to ^{239}Pu in terms of their specific contributions to the count rate. $^{239}\mathrm{Pu}_{\mathrm{eff}}$ can also be considered as the "fissile content equivalent" of an SFA. The subscript "CIPN" stands for "CIPN technique" and is needed to distinguish the $^{239}\mathrm{Pu}_{\mathrm{eff}}$ determined with a CIPN instrument from that determined by another instrument. The subscript "m" stands for mass of each isotope.

Besides the aforementioned 3 fissile isotopes. ^{238}U also produces neutrons, but its contribution is sufficiently depressed in water and its mass changes slowly with burnup. Besides, since ^{238}U has a $\sim$1 MeV fission cross section threshold, the majority of the ^{238}U fission is caused by the fission neutrons of the three aforementioned fissile isotopes, given only a small fraction of ^{238}U directly interact with Cf source neutrons. So most of the ^{238}U fission ($\sim$90%) can be "tied" to the multiplication of the 3 main fissile isotopes. The neutron contribution from ^{238}U fission among all SFAs were also quantified. Eq. 6.1 shows the fraction of neutron produced by ^{238}U in all

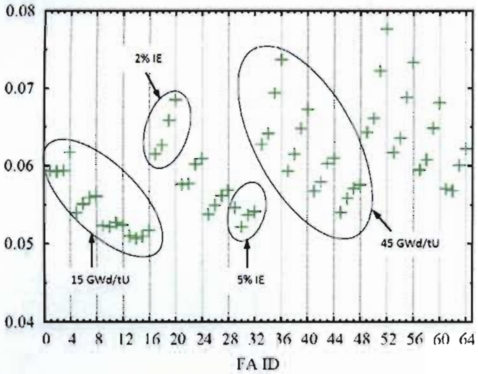


Fig. 6: The fraction of neutron produced by ^{238}U in all 64 SFAs (with the ID of each SFA goes from 1 to 64). The FA ID is ordered by CT− >IE− >BU.

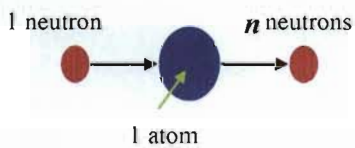


Fig. 7: Diagram of the "net neutron contribution" concept.

64 SFAs. The ID of SFA is ordered by CT− >IE− >BU, e.g., FA with ID 1, 2, 3 and 4 means assembly with BU of 15GWd/tU, IE of 2%, CT of 1, 5, 20 and 80 years, respectively. The neutron contribution of ^{238}U ranges from 5% to 7.5% with the average around 6%. Since the contribution from ^{238}U is relatively small and varies in a small range, as well as most ^{238}U fission can be attributed to the multiplication of the three main fissile isotopes, ^{238}U can be treated as a background factor.

6.2. "Net Neutron Contribution" concept used to determine the weighting coefficient C_i

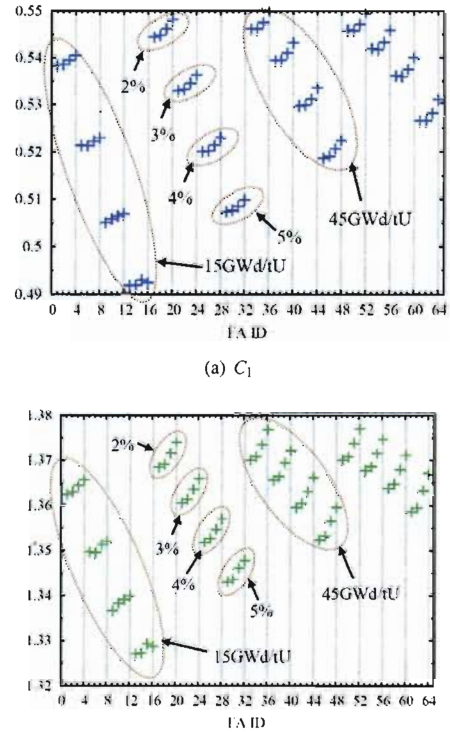
A useful concept for quantifying the relative impact of various isotopes on neutron balance in spent fuel is the concept of "net neutron contribution". Fig. 7 shows a diagram of this concept. Suppose a neutron collide with an atom of certain isotope (e.g., ^{235}U, ^{239}Pu, etc.), and the statistical average number of out-coming neutrons are assumed to be "n". The weight of one particular isotope in terms of "net neutron contribution" is proportional to "n". Eq. 3 shows the expression of "n", which is the "net gain" between the different number of neutrons produced by fission and lost by absorption, where: σ_f and σ_a are fission and absorption cross section, respectively; ν is average number of neutrons produced per fission.

$$n \propto (\overline{\nu}-1)\overline{\sigma_f}-\overline{\sigma_a}; \text{where:} \tag{2}$$

$$\sigma_a=\sigma(n,\gamma)+\sigma(n,\alpha)+\cdots;\ \sigma_a \text{ does not include } \sigma_f. \tag{3}$$

In the context of spent fuel, σ_f, σ_a and ν are all energy-dependent, while the neutron flux, Φ, are both energy- and space-dependent. C_i is the "weight" of one particular isotope relative to that of ^{239}Pu. The full expression of C_i is shown below:

$$C_i=\frac{\int_V\int_E \sigma_f(E)_i(\nu_i(E)-1)\Phi(E,V)dEdV-\int_V\int_E \sigma_a(E)_i\Phi(E,V)dEdV}{\left[\int_V\int_E \sigma_f(E)(\nu(E)-1)\Phi(E,V)dEdV-\int_V\int_E \sigma_a(E)\Phi(E,V)dEdV\right]_{^{239}Pu}}, \tag{4}$$



(a) C_1

(b) C_2

Fig. 8: value of weighting coefficient (C_i) for each of the 64 FAs: (a) C_1; (b) C_2.

With Eq. 4, C_1 ($i=1$) can be obtained by plugging in relevant parameters of ^{235}U in the numerator, and C_2 ($i=2$) can also be obtained for ^{241}Pu. This equation can be implemented using flux multiplication tallies in MCNPX. The second term in the numerator can be calculated using neutron flux tally ("F4") over the entire fuel region multiplied with "FM -2" (absorption multiplication tally). The same flux tally can then be multiplied with "FM -6 -7" and "FM -6" (fission neutron rate and fission rate, respectively), and the difference of these two tallies results in the first term in the numerator. Similar approach can be used for the denominator. Note that from this equation, C_1 and C_2 are obtained on "per atom" base; to convert to "per gram" base, the molar mass ratios of "239/235" and "239/241" are multiplied respectively to the value of C_1 and C_2 determined by this equation. Fig. 8 shows the values of C_1 and C_2 for each of the 64 SFAs (with FA ID goes from 1 to 64 in the same order as in Eq. 6.1). It can be observed from these two figures that there is a clear trend of C_1 and C_2: higher IE, lower values for both C_1 and C_2. It also shows that C_1 and C_2 are dependent on BU and CT as well, but not as prominent as on IE. In general, for the same BU and CT, the neutron energy spectrum in assembly with higher IE have lower thermal neutron peak. Also the neutron energy spectrum in the fuel is "shaped" by the presence of many isotopes, e.g., the unique resonant absorption of a particular isotope will depress the neutron flux in its resonant energy range. These two factors are suspected to contribute to the trend observed in C_1 and

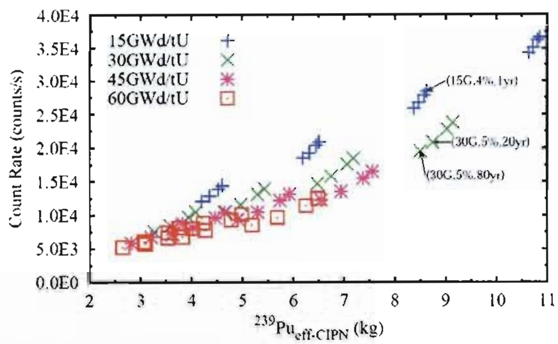


Fig. 9: Count rate (counts/sec) vs. $^{239}Pu_{eff\_CIPN}$ (gram) for all 64 SFAs. The count rate is normalized with a 100-μg ^{252}Cf source, which releases $2.34E8$ neutrons/sec. Each SFA represents a unique combination of burnup (BU) (15, 30, 45 and 60 GWd/tU), initial enrichments (IE) (2, 3, 4 and 5%) and cooling time (CT) (1, 5, 20 and 80 years). Each color represents one BU level. For the 16 FAs with the same BU, four groups of data points are shown. And within each group, the 4 FAs have the same IE but different CT.

C_2. The average value of C_1 and C_2 ($\overline{C_1}$ and $\overline{C_2}$ respectively) are 0.529 and 1.359, respectively. The value of C_1 fluctuates from the average value in the range of (-7%, 4%); and the value of C_2 fluctuates in a smaller range of (-2%, 1%).

6.3. Count Rate vs. $^{239}Pu_{eff\_CIPN}$

The count rate above background (net count rate) for each of the 64 SFAs were obtained using the MCNPX simulations. The mass of ^{235}U, ^{241}Pu and ^{241}Pu of each assembly were obtained from the NGSI virtual library, which are generated using burnup calculations. And $^{239}Pu_{\text{eff\_CIPN}}$ was then calculated using Eq. 1. $\overline{C_1}$ and $\overline{C_2}$ from the previous section were used in this calculation.

Fig. 9 shows net count rate as a function of $^{239}Pu_{\text{eff\_CIPN}}$ for all the 64 SFAs without any corrections. Each color represents one BU level. For the 16 assemblies with the same BU, four groups of data points are shown; one group represents each of the 4 IEs. And within each group of 4 assemblies with the same BU and IE, there are 4 assemblies with different CT. As shown, the data points are scattered, and there is NO coherent relation between count rate and $^{239}Pu_{\text{eff\_CIPN}}$ (i.e., given a count rate, a unique quantity of $^{239}Pu_{\text{eff\_CIPN}}$ can not be determined).

An ideal outcome from Fig. 9 would have the count rate of all 64 SFAs be a simple function of $^{239}Pu_{\text{eff\_CIPN}}$ mass. This would indicate that Eq. 1 using $\overline{C_1}$ and $\overline{C_2}$ properly weights the relative significance of the three fissile isotopes and that there are not any other significant factors impacting the count rate beside the three primary fissile isotopes. But that is not the reality. The main conclusions from Fig. 9 are the following: 1) The count rate among the SFAs generally trend with the $^{239}Pu_{\text{eff\_CIPN}}$. 2) There is structure in the data indicating that the count rate is a function of factors that scale with BU, IE and CT. The strongest dependence is a function of BU. That is $^{239}Pu_{\text{eff\_CIPN}}$ is not the only influencing factor; other factors such as neutron absorbers must also at play.

6.4. Count Rate vs. $^{239}Pu_{eff\_CIPN}$ with Corrections

The two primary factors most likely to impede a smooth relationship between count rate and $^{239}Pu_{\text{eff\_CIPN}}$ are (1) neutron absorption and (2) neutron production from isotopes besides the three main fissile isotopes. Both factors were investigated. Regarding the second factor, ^{238}U is a main neutron creator besides ^{235}U, ^{239}Pu and ^{241}Pu. As discussed earlier, ^{238}U can be treated as a background component in the medium of water.

Two kinds of neutron absorbers play major role in terms of neutron absorption in an SFA: (a) actinide neutron absorbers (e.g., ^{240}Pu) and (b) fission fragment absorbers (e.g., ^{149}Sm). Among these absorbers, most of them accumulate with burnup because the longer the fuel burns, the more actinides and fission fragments accumulate. Two notable exceptions are ^{155}Gd and ^{241}Am; they both grow dramatically with cooling time. ^{155}Gd is a decay daughter of ^{155}Eu with a half-life of 4.68 years, and ^{155}Gd has extremely large neutron absorption cross section. ^{241}Am is a decay daughter of ^{241}Pu with a half-life of 14.4 years, and ^{241}Am has large neutron absorption cross section as well (and only a small fission cross section).

In order to include the impact of neutron absorbers, BU and CT corrections were introduced to $^{239}Pu_{\text{eff\_CIPN}}$ based on their dependence on BU and CT. And these two corrections change $^{239}Pu_{\text{eff\_CIPN}}$ to "$^{239}Pu_{\text{eff\_CIPN}}$ with corrections" (also indicated by "X"). The BU and CT corrections are expressed as below:

$$X \equiv {}^{239}\text{Pu}_{\text{eff\_CIPN}} \text{ with corrections} = C_{BU}\left[{}^{239}\text{Pu}_{\text{eff\_CIPN}} + f(BU,CT)\right] \quad (5)$$

$$\text{or, } {}^{239}Pu_{eff\_CIPN} = X/C_{BU} - f(BU,CT) \quad (6)$$

where C_{BU} (BU correction coefficient) is used to account for the absorption from the fission fragment absorbers and actinide absorbers scale with BU. C_{BU} can be quantified by weighting the "absorbing power" of these absorbers at higher BU relative to 15 GWd/tU. The "absorbing power" can be calculated by lump-summing all these stable absorbers with proper weighting of each isotope in each FA with different BU. With the results of C_{BU} at different BU, an empirical relation of C_{BU} was established by using power-law fitting, as shown below:

$$C_{BU} = (BU/15)^{-0.302}, \text{ for } 15 \leq BU \leq 60 \text{ GWd/tU} \quad (7)$$

As discussed before, two major neutron absorbers are found to change dramatically with CT: ^{155}Gd and ^{241}Am. So the CT correction, $f(BU,CT)$, can be written as:

$$f(BU,CT) = C_3{}^{155}\text{Gd}_m + C_4{}^{241}\text{Am}_m \quad (8)$$

As shown in Eq. 8, the CT correction can be expressed as weighted sum of ^{155}Gd and ^{241}Am. The value of C_3 and C_4 can be obtained similarly as C_1 and C_2 by using Eq. 4. Since both isotopes have negligible fission cross section, the value of C_3 and C_4 are both negative. The average value of C_3 and C_4 are -48.97 and -0.66 respectively. With these values, together with Eq. 7 and Eq. 8, Eq. 5 now becomes:

$$X = \left(\frac{BU}{15}\right)^{-0.302}\left[{}^{239}\text{Pu}_{\text{eff\_CIPN}} - 48.97{}^{155}\text{Gd}_m - 0.66{}^{241}\text{Am}_m\right] \quad (9)$$

With the equation above, the "X" value ("$^{239}Pu_{\text{eff\_CIPN}}$ with corrections") can be easily calculated, with the masses of the 5 isotopes (i.e., ^{235}U, ^{239}Pu, ^{241}Pu, ^{155}Gd and ^{241}Am) in a particular SFA obtained through burnup calculations. And then

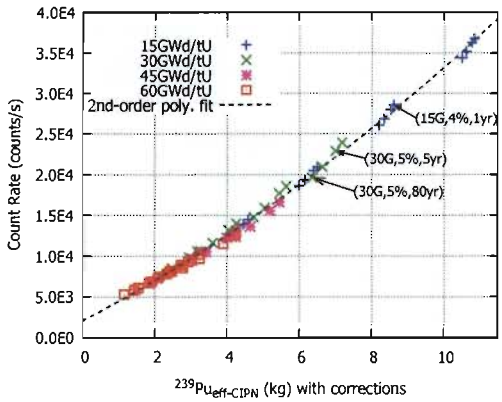


Fig. 10: Count rate (counts/sec) vs. "^{239}Pu$_{eff_CIPN}$ with corrections" (X) (kg) for all the 64 SFAs. BU and CT corrections have been introduced to ^{239}Pu$_{eff_CIPN}$.

value of "X" and count rate (CR) for each of the 64 SFAs is plotted in Fig. 10. Fig. 10 illustrates the relation between count rate (CR) and "X". As shown, a coherent universal relation between CR and "X" exists (i.e., given a certain count rate, the value of "X" can be uniquely determined, and vice versa). Note that CR does not reduce to zero when "X" goes to zero because of the Cf background, ^{238}U background, etc. Also indicated by dash line in this figure, the fitting curve agrees with the data points quite well. With the 2nd-order polynomial fitting, the empirical relation between CR and "X" is expressed as below:

$$CR = 80.65X^2 + 2307.4X + 2029.68 \qquad (10)$$

The standard deviation of the relative difference between the real CR and the predicted CR by this fitting function is 3.2%.

From a practical point of view, for an unknown SFA, once the CIPN count rate is measured, "X" can be uniquely determined through Fig. 10 (a calibrated version). The fissile content, ^{239}Pu$_{eff_CIPN}$, can then be calculated using Eq. 9, provided BU and the mass of ^{241}Am and ^{155}Gd can be quantified. Usually, the BU of an SFA can be determined using the measured gross gamma emission obtained in a passive gamma technique [10], and the CT can be verified based on the operator's declarations. But the mass of ^{241}Am and ^{155}Gd are usually unknown. Since the production path of both ^{241}Pu and ^{155}Gd are relatively simple, empirical relations can be established by using decay functions and by fitting existing data from burnup calculations. These relations can then be used to predict the mass of ^{241}Am and ^{155}Gd as function of BU and CT [11]. An alternative is to measure the characteristic gamma lines of ^{241}Am and ^{155}Eu (or ^{154}Eu) obtained in a gamma spectrometry technique [12, 13]. Future work is needed to further study both options.

7. Conceptual Experiment Setup

The conceptual experiment setup of CIPN has been proposed and presented here in Fig. 11. Compared to the CIPN design shown in Fig. 2(a), a "door" (made of polyethylene) has been added to hold the ^{252}Cf source on the front side, and the two arms are extended to support the door, as shown in Fig. 11. Also an ion chamber is added to each arm of the fork, in addition to the existing fission chambers. The ion chamber will be used to measure gross gamma emission. As we

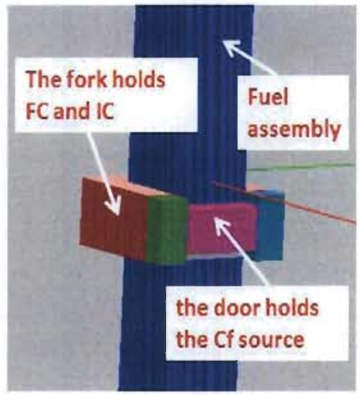


Fig. 11: The conceptual experiment setup of the CIPN instrument

know, the Fork detector has been used in the field for decades and Fig. 12 shows a typical Fork measurement setup. Since CIPN is like a Fork detector, the setup of the body of CIPN is nearly identical except the arms of the fork will extend further, beyond the dimensions of the fuel, so the source door can be attached. The source door will look like the one in a Collar detector, as shown in Fig. 13(a), so that the door can slide in and out of the detector through the groove in the detector body. A similar "L"-shaped pole as shown in Fig. 12 will be used to hold the door. Fig. 13(b) shows the top view of the door and the hole in the top is used to guide through the Cf source into position. The design of CIPN with a detachable door can avoid the possibility of getting the instrument stuck in a bowed fuel assembly.

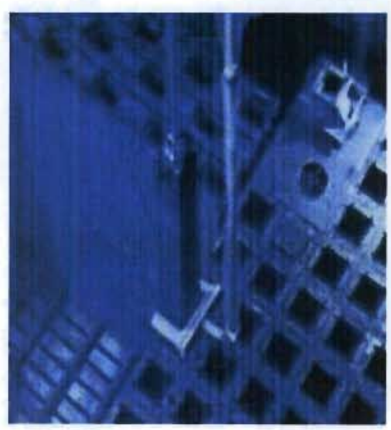
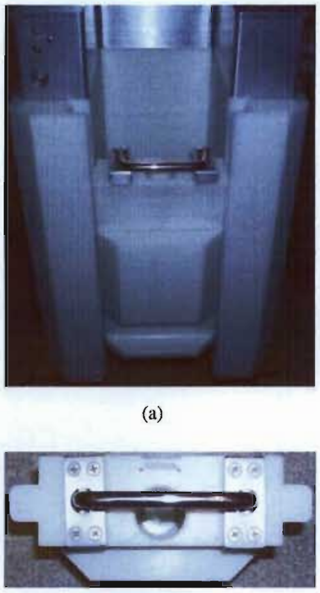


Fig. 12: The experiment setup for the Fork detector



(a)

(b)

Fig. 13: The Collar detector for fresh fuel assemblies. (a) Front view of the Collar detector; (a) Top view of the door of the Collar detector.

Summary

In summary, a new neutron detector, CIPN, was proposed to quantify fissile content in spent fuel assemblies. The capability of CIPN has been quantified using a series of virtual spent fuel assemblies and encouraging results have been obtained. The conceptual experiment setup of CIPN is also briefly discussed at the end. CIPN is sensitive to diversion and it has almost uniform response to diversion at different locations across the assembly. With the schemes presented in this paper, together with given (either measured or verified) BU and CT, the fissile content of a target spent FA (or $^{239}Pu_{eff\_CIPN}$) can be determined within a few percent. The precision is excellent with less than 1% statistical uncertainty obtained in less than 100 second. The accuracy will likely be limited by the calibration and systematic errors. The neutron source needs to be $\sim 2 \times 10^8$ n/s which corresponds to a ^{252}Cf source of 100 μg. It was demonstrated that burnup and cooling time corrections are needed to accurately predict the fissile content of a given assembly, although further research is needed to determine some of the important neutron absorbers, such as ^{155}Gd and ^{241}Am, either by empirical relations or by measurement using other techniques. Integrating CIPN with other NDA techniques to measure plutonium in spent fuel assemblies are also under way. Fabrication of the CIPN instrument is planned for 2012 and measurement of spent fuel of CIPN is expected in 2013.

Acknowledgements

The authors would like to acknowledge the support of the Next Generation Safeguards Initiative (NGSI), Office of Nonproliferation and International Security (NIS), National Nuclear Security Administration (NNSA).

The authors would also like to thank Dr. Martyn Swinhoe, Dr. Stephen Croft, Dr. Michael Fensin, Dr. John Hendricks, Dr. Taehoon Lee, Dr. Jeremy Conlin and Ms. Melissa Schear for their valuable inputs to this project.

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