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Results of the Swedish Spent Fuel Measurement Field Trials with the Differential Die-Away Self-Interrogation Instrument

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Abstract

Differential Die-Away Self-Interrogation (DDSI) is a method by which the characteristic die-away of neutrons from spontaneous and induced fissions is used to characterize a spent nuclear fuel assembly. A nondestructive assay (NDA) instrument was built at Los Alamos National Laboratory to implement and test the DDSI method. The DDSI instrument contains 3He detectors which measure thermal neutrons, and the time and location of detection of each neutron is recorded via list-mode data acquisition. The instrument was sent to the Clab interim storage facility in Sweden for measurement and characterization of 50 spent pressurized water reactor (PWR) and boiling water reactor (BWR) fuel assemblies. The result was over 40 hours of neutron list-mode data from a wide variety of fuel assemblies with high enough efficiency to perform neutron coincidence counting, i.e. detection of time-correlated neutrons from fission. Analysis algorithms for characterization of the fuel assemblies were tested on the Swedish spent fuel dataset. Using the measured data, multiplication, fissile mass, initial enrichment, burnup, and total plutonium mass were determined in the 50 assemblies with root mean square errors ranging from 1.5% for PWR multiplication to 11.4% for BWR fissile mass. The results in this work demonstrate that the DDSI concept is capable of characterizing spent power reactor fuel with levels of accuracy that are compatible with the requirements and objectives of various applications such as safeguards verification or facility material control and accounting.

1. Introduction

Differential Die-Away Self-Interrogation (DDSI) is a method by which the characteristic die-away of neutrons from spontaneous and induced fissions is used to characterize a spent nuclear fuel assembly. A nondestructive assay (NDA) instrument based on the DDSI method was developed at Los Alamos National Laboratory (LANL) in 2009 [1] as a part of what was then the Next Generation Safeguards Initiative (NGSI) project [2]. The DDSI instrument, henceforth referred to simply as DDSI, is the first spent nuclear power reactor fuel NDA instrument capable of performing neutron list mode data collection and coincidence counting. DDSI was simulated extensively and several analysis algorithms were developed in the following years [3]. Fabrication of the DDSI took place in 2014, after which it was used to characterize fresh fuel at LANL [4]. The project culminated in a five week spent fuel assembly measurement campaign at the Clab interim storage facility in Sweden, the results of which are presented in this paper.

A total of 50 assemblies, 25 Pressurized Water Reactor (PWR) and 25 Boiling Water Reactor (BWR) type, were measured in Sweden with a static, central measurement and with an axial scan. The technical goals of the project, and therefore of the field trials at Clab, were to: (1) verify the initial enrichment, burnup, and cooling time of facility declaration, (2) detect the diversion or replacement of pins, (3) estimate fissile mass, (4) estimate decay heat, and (5) determine the reactivity of spent fuel assemblies.

2. Overview of DDSI Method and Instrument

The DDSI method relies on the fact that spontaneous fission and (α,n) neutrons, primarily from ^{244}Cm in spent nuclear power reactor fuel, thermalize in the water between pins and induce fissions in the fissile material in the assembly [1]. By measuring time-correlated neutrons from spontaneous and induced fissions, nuclear material can be characterized [5].

Neutrons are detected using 28 ^{3}He tubes on two sides of a spent fuel assembly. The time of detection of every neutron is recorded (known as list-mode data collection) together with the detector channel allowing also for spatial reconstruction [6].

The DDSI instrument contains four stainless steel pods, two of which were used for this measurement campaign (pods 2 and 3) while the other two were not able to be used due to mechanical design-related issues (pods 1 and 4). Two pods were determined to be sufficient for showing the usefulness of the DDSI method. Each pod contains a block of high-density polyethylene with 15 cylindrical slots, each with space for a 1-inch-diameter ^{3}He tube. Fourteen of the slots in each pod hosted a ^{3}He tube and contributed to the DDSI signal. The remaining slot in pod 2 was used for an ion chamber for gross gamma-ray counting and the remaining slot in pod 3 was used for a ^{3}He tube with a newly developed KM200 preamplifier [7] (analysis and discussion of the ion chamber and ^{3}He tube with KM200 preamp results exceeds the scope of this paper). Each DDSI ^{3}He tube had its own dedicated preamplifier, the signal from which was run through Tygon tubing up to the main data acquisition computer (DAQ) located by the side of the pool. Extensive details about the geometry of the DDSI instrument can be found in [3]. Figure 1 contains a top-down schematic of the DDSI instrument, showing which channels and pods were used in these field trials.

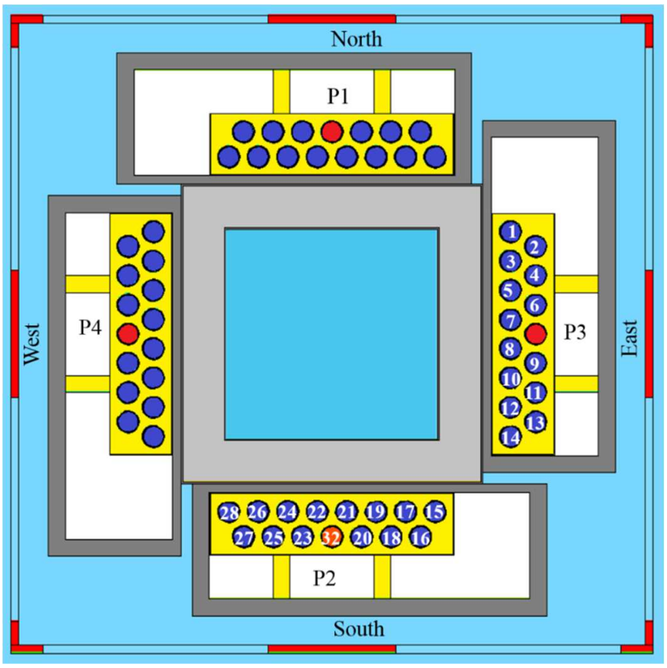

Figure 1. Schematic of the DDSI instrument showing orientation in the facility and pods used in the field trials (P2 and P3) with output channels labeled.

3. Experiment Details

Clab spent fuel assemblies were manipulated by specially designed fuel handling equipment attached to a crane and operated by Clab personnel. Each assembly was lifted from its storage container, moved laterally to be positioned above the DDSI fuel funnel opening, and then lowered into a central position for a 20 or 30-minute static measurement. Figure 2 shows a PWR assembly in the central position of the DDSI instrument. The assembly was then lowered as far as the crane could move it (which left a portion of the assembly above the funnel opening) and then slowly raised out of the instrument for the axial scan measurement. Figure 3 shows a schematic of the positioning of PWR assemblies in DDSI at the center and lowest positions, and Figure 4 shows the same schematic for BWR assemblies. The PWR assemblies have visible pins, which allowed for a reasonable approximation of the active region of the fuel; however, the BWR assemblies are covered with a sheath, which made it difficult to identify where the active region actually began and ended. Therefore, for repeatability, all measurement positions are reported in terms of how much has the fuel assembly been lowered, which is a value provided and recorded by the Clab assembly handling equipment. During the static center measurements, the true active center of the PWR and BWR fuel assemblies were 647 mm and 821 mm, respectively, below the active center of the ^{3}He tubes.

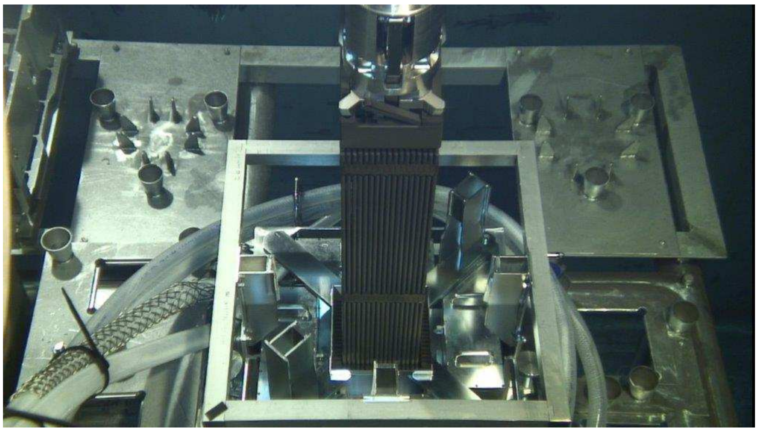

Figure 2. PWR fuel assembly suspended in the DDSI instrument underwater at the Clab facility in Sweden.

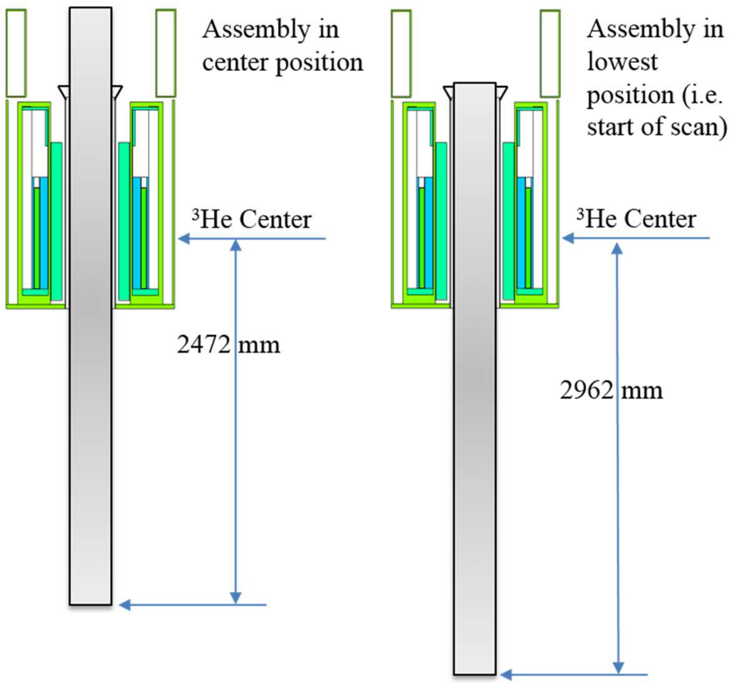

Figure 3. Schematics showing positioning of PWR assemblies in DDSI for the static center measurement (left) and the lowest position where the axial scan starts (right).

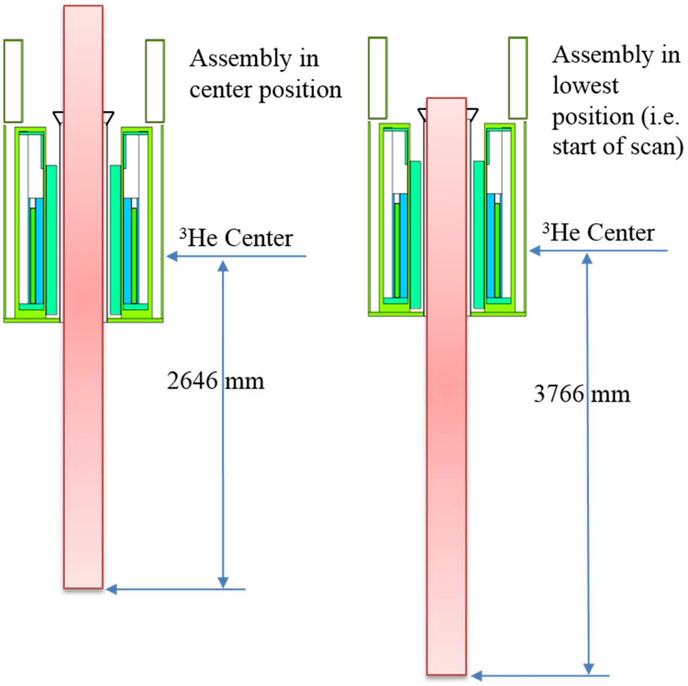

Figure 4. Schematics showing positioning of BWR assemblies in DDSI for the static center measurement (left) and the lowest position where the axial scan starts (right).

The active region of fuel in the BWR assemblies can be lowered further into the funnel than in the PWR assemblies because of the geometric differences.

Comparing the count rates in individual channels (i.e. individual ^{3}He tubes) during the measurements provided insight into how the assemblies were positioned radially. The PWR and BWR assemblies contain different numbers of pins in matrices of different cross sectional area, therefore some assemblies have more space between the pins and the funnel edge than others. One of the benefits of list-mode data collection (recording the time of arrival of neutrons in every channel) is that the channel count distributions can be used to derive radial positioning information. This is discussed in detail in [8].

3.1. Data acquisition and analysis

The signals from 28 ^{3}He tubes from two pods (14 tubes per pod) were transmitted into the data acquisition (DAQ) system via a specialized input card designed and built at LANL. The National Instruments List-Mode Acquisition (NILA) software on the DAQ recorded neutron timing information in list mode, i.e. the time and channel of every detection event was recorded. The static center measurements were analyzed by converting the list-mode neutron data into Rossi-alpha Distributions (RADs) [2]. An optimized RAD production algorithm was implemented into the FastTapX software [9] in a special version made specifically for DDSI and Differential Die-Away (DDA) data analysis, referred to as FastTapX-DDSI [10]. The software was used to produce and store a unique RAD for each of the more than 150 static measurements taken.

Axial scan data, both for neutrons and gammas, were analyzed by calculating the singles count rate using the “Get Rates” function of FastTapX-DDSI in every file recorded (note: by default, each file was 5 s long). Each 5 s data point corresponded to approximately 18 mm of fuel movement. Each 5 s measurement could therefore be associated with an axial measurement location on the assembly, and analysis of count rate as a function of axial height on the assembly could be performed. In this way, BWR assemblies with uneven axial burnup distributions were corrected such that the reported burnup was the approximate burnup at the location of the long static measurement, as opposed to the average burnup of the entire fuel assembly length. This value is henceforth referred to as “adjusted burnup”, or “ABU”. PWR assemblies have fairly even axial burnup distributions and therefore a correction was not necessary.

3.2. Operating voltage

Due to different irradiation history of individual fuel assemblies, the ratio of neutron and gamma source terms may significantly vary between different fuel assemblies. Hence it is of critical importance to avoid contribution of gamma pileup to the measured ^{3}He count rates, as this may lead to a variable enhancement of the measured rates that is not correlated to the actual strength of the neutron source term. In order to evaluate the magnitude of the potential gamma pileup issue, we have measured high voltage plateaus with the highest and the lowest count rate PWR and BWR assemblies, shown in Figure 5. Full plateaus were measured with the shift register-based electronics to assess the gamma pileup in the system as a whole. The results show that operating at 1600 V leads to very little gamma pileup, only of about 1-3% between the least and the most active fuel assemblies, while operating at 1680V results in potential contribution of gamma pile up of the order of 3-10%. Therefore, 1600 V was chosen as the operating HV instead of 1680V which is traditionally associated with ^{3}He detector tube design used in DDSI. The loss in efficiency due to the lower operating HV was quantified by taking a ratio of singles from ^{252}Cf (which does not produce significant number of gammas and hence does not lead to gamma pileup) at 1680V and 1600V and determining the fractional loss in singles counts. The resulting fractional loss in singles counts was ~11%, and the loss in doubles counts was ~23% which is considered to be statistically less significant than distortion of measured neutron counting rates due to changing contribution of the gamma pileup.

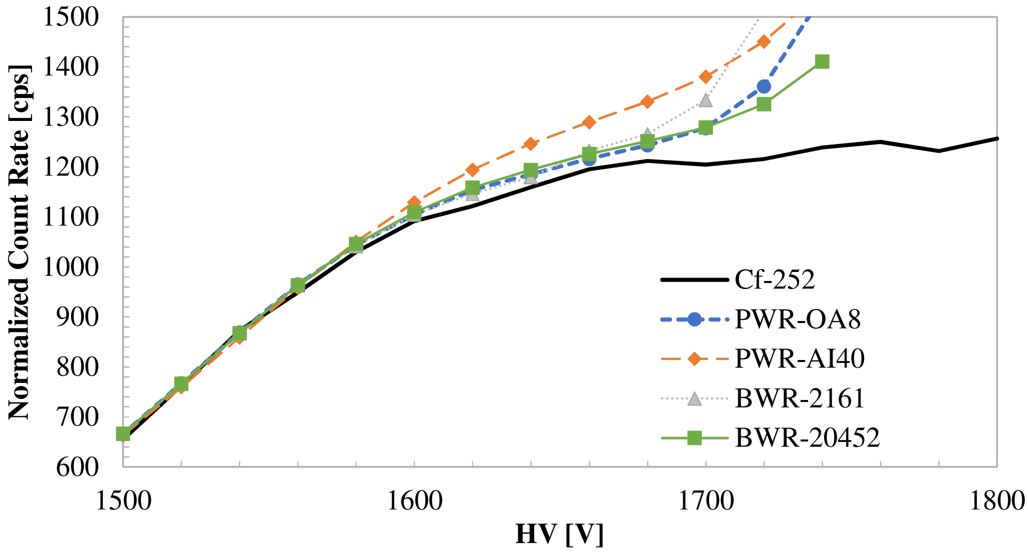

Figure 5. Normalized high voltage plateaus from two PWR and two BWR assemblies compared to ^{252}Cf plateau.

By examining HV plateaus on a channel-by-channel basis, the gain of channel 9 (located in Pod 3) was found to have drifted prior to the measurement by a factor compatible with HV change of approximately 30V and therefore was removed from the analysis. The unique list-mode capabilities of DDSI enabled detection and elimination of this malfunctioning channel.

3.3 Spent Fuel

The set of PWR and BWR spent fuel assemblies measured in this campaign are collectively referred to as the "SKB-50". The SKB-50 assemblies come from 12 commercial reactors operated in Sweden, resulting in a diverse set of fuels with a wide range of initial enrichments, burnups, and cooling times [11]. Of the PWR assemblies, five comprise a 15 × 15 array of pins, and 20 PWR comprise a 17 × 17 array. In the set of BWR assemblies, 11 are 8 × 8 arrays of pins, and 14 have 10 × 10 arrays of pins. This diversity provides an opportunity to test the effectiveness of characterization algorithms in the extremes, such as assemblies with strong axial or radial burnup distributions, underburned assemblies, and assemblies with different geometries. One characteristic that largely encompasses the effects of these varying parameters is assembly net multiplication, defined as neutrons produced per source neutron and calculated using simulated models of the fuel assemblies in MCNP [12]. Table I provides the net multiplication values for the SKB-50 fuel assemblies. The PWR multiplication values are generally higher than the BWRs, and represent a slightly larger range. The standard deviation of net multiplication is larger for the BWRs, presumably because during irradiation, some axial positions at the bottom of each assembly are moderated by water whereas others at the top of the assembly are moderated by steam, leading to significant isotopic variation along the axis of each individual fuel assembly.

Table I. Net multiplication values for the SKB-50 fuel assemblies.

	Net Multiplication	
Assembly Number	**PWRs**	**BWRs**
1	1.640	1.317
2	1.666	1.329
3	1.575	1.329
4	1.675	1.335
5	1.675	1.335
6	1.580	1.270
7	1.704	1.344
8	1.521	1.345
9	1.663	1.349
10	1.624	1.368
11	1.605	1.297
12	1.533	1.384
13	1.556	1.363
14	1.623	1.309
15	1.492	1.304
16	1.645	1.327
17	1.671	1.296
18	1.620	1.365
19	1.628	1.343
20	1.623	1.460
21	1.623	1.331
22	1.604	1.527
23	1.844	1.592
24	1.595	1.383
25	1.640	1.403

4. DDSI Experimental Results

Background, characterization, and fuel measurements were performed in a Clab testing pool where spent fuel may temporarily reside. Background was measured to ensure that the neutron signal from assemblies in storage waiting to be measured did not affect our measurement signal. The results of these measurements show that the background was not statistically significant no matter what the fuel assembly configuration was in the external fuel container, shown in Figure 6. Characterization measurements with a ^{252}Cf source were performed prior to the first fuel assembly measurement to ensure that all detectors were functioning correctly and that the die-away time derived from the results was similar to the value expected based on previous measurements at LANL. The instrument die-away time of 18 μs measured with ^{252}Cf was indeed consistent with previous measurements at LANL reported in earlier work [3].

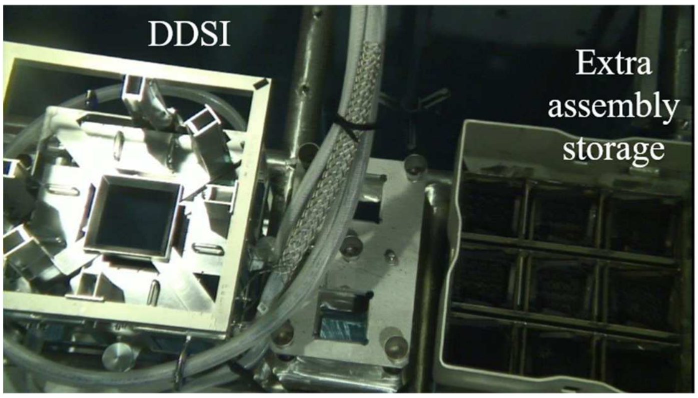

Figure 6. Image of DDSI in the pool showing the instrument location relative to the extra assembly storage container.

4.1. Early die-away time analysis

Net multiplication is defined as the number of neutrons produced per source neutron, i.e. neutron released from spontaneous fission of a nucleus inside the pins of the spent fuel assembly. This quantity is an inherent indication of the balance between fissile material and the neutron absorbers in a fuel assembly. Rossi-alpha distributions (RADs) are histograms of times between neutron captures and the shapes of the distributions are correlated with assembly characteristics, including multiplication [3]. The early die-away time (τ_e) method [13] was applied to experimentally-obtained RADs to determine net multiplication in both the PWR and BWR assemblies. Using the early die-away method, multiplication estimates in the PWR assemblies have an RMS error of 1.5%, and BWR assemblies have an RMS error of 2.2%, as reported in detail in [8]. For completeness, Figure 7 from [8] is reproduced here.

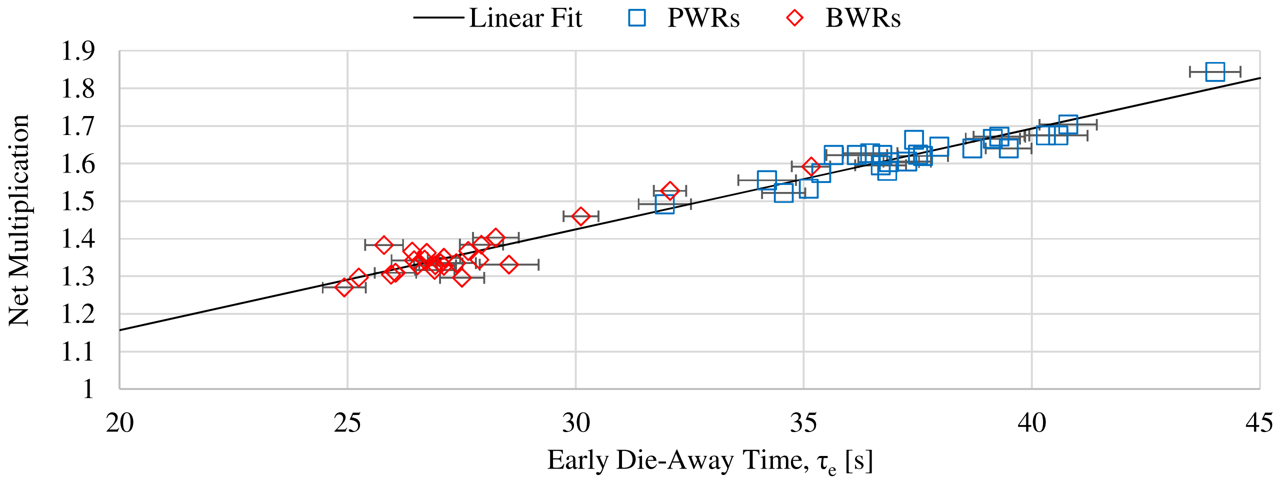

Figure 7. Early die-away time versus net multiplication for SKB-50 assemblies, reprinted from [8] .

4.2 Total plutonium mass

The total mass of plutonium in a spent fuel assembly is of interest for criticality safety, nuclear material accountancy, and potentially, safeguards measurements. A method of plutonium mass determination in spent power reactor fuel assemblies was developed and demonstrated using simulated Differential Die-Away nondestructive assay (NDA) measurements in 2013 [14]. The method was then adapted for the Differential Die-Away Self-Interrogation instrument in 2014 by using the early die-away time as a measure of multiplication [15] and determining Pu mass in different simulated assemblies [16]. The expression developed for plutonium mass in this work is given in equation (1):

$$m_{Pu} = d(CT)\left(\frac{\tau_e + g}{\tau_e + h}\right)\left(\frac{S}{\tau_e + g}\right)^{f(CT)} \tag{1}$$

where m_{Pu}= plutonium mass, CT = cooling time, τ_e = early die-away time, S = singles, and d, f, g and h are the calibration parameters which depend on the instrument design, and in the case of d and f, also on CT. This expression was successful in determining plutonium mass in the simulated assemblies with a root mean square error of 2.1% across Spent Fuel Library 2a [17]. This assumed that cooling time could be taken as a known quantity (be it from trusted declaration or from experimental measurements such as passive gamma measurements), but was independent of initial enrichment and burnup. The calibration constants d and f were functions of cooling time.

This method was applied to the spent fuel data collected at Clab. We found that the calibration differed from previous work in that the results were independent of cooling time. This was at first rather surprising because it is often unusual for experimental data to follow more simplified trends than simulated data. This can be explained, however, by considering the range of fuel assembly configurations simulated vs measured. In simulation space, we included a wide range of IE, BU, and CT combinations, excluding only those that were impossible, not those that were improbable. The set of measured SKB-50 assemblies, while still representing a wide range of irradiation histories, contains nearly exclusively assemblies burned in a more practical way with focus on the economics of commercial power generation. Figure 8 shows the range of IE, BU, and CT conditions simulated vs. measured. These plots clearly demonstrate that in the simulations we were oversampling parameter space that included unrealistic assemblies that are not represented in the SKB50 set. Regardless, the measured data follow smooth trends independent of cooling time as observed in Figure 9 and Figure 10.

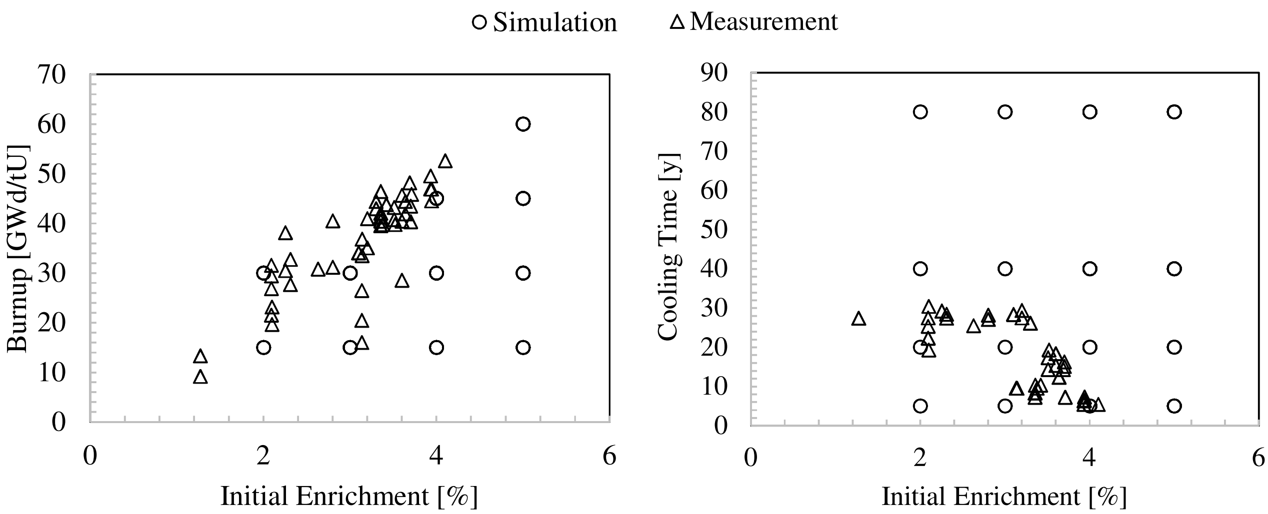

Figure 8. BU vs. IE and CT vs. IE for measured and simulated assemblies to demonstrate the range of assembly configurations analyzed.

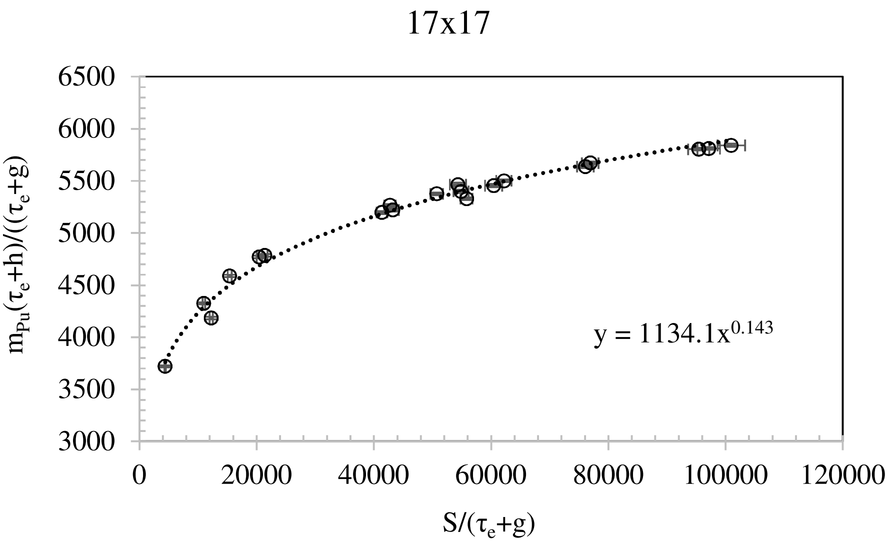

Figure 9. Power function relation between plutonium mass and singles rate, weighted by early die-away time and calibration constants for 17x17 assemblies.

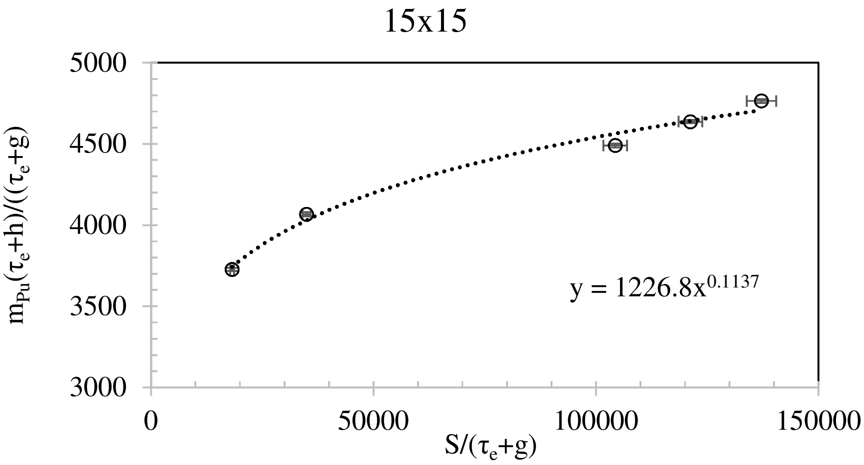

Figure 10. Power function relation between plutonium mass and singles rate, weighted by early die-away time and calibration constants for 15x15 assemblies.

Using these relations, total plutonium mass is determined in 17x17 and 15x15 assemblies with root mean square errors of 1.8% and 1.1%, respectively. When all assemblies are combined into a single calibration curve, total plutonium mass is determined with a RMS error of 2.0% as shown in Figure 11. In another departure from trends observed in simulation space, the data are slightly better represented by a logarithmic function, which results in an equation for total Pu that takes the form shown in equation (2):

$$m_{pu} = \left(\frac{\tau_e + g}{\tau_e + h}\right)\left[d * ln\left(\frac{S}{\tau_e + g}\right) - f\right] \tag{2}$$

where the variables are the same as in equation (1). When the logarithmic fit is used instead of the power function, the RMS error of the PWR dataset as a whole drops from 2.0% to 1.7%. The RMS error for 15x15 assemblies does not change when the logarithmic fit is applied, and it drops from 1.8% to 1.6% for the 17x17 geometry.

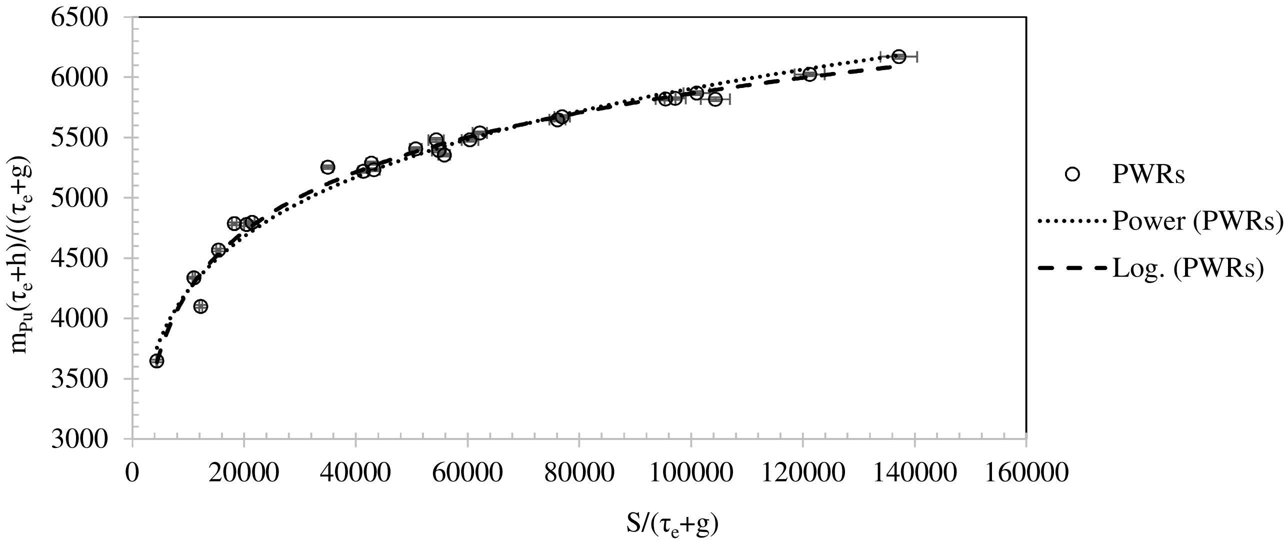

Figure 11. Power and logarithmic function relations between plutonium mass and singles rate, weighted by early die-away time and calibration constants for all PWR assemblies.

The same method was successfully applied to the BWR assemblies as shown in Figure 12. The fit is not as consistent as with the PWRs because of the wide variation in axial burnup distribution in the BWRs. The total plutonium value encompasses the entire assembly, whereas the central measurement only captures a small portion of the axial length. Regardless, the total plutonium determination method applies well to the BWR dataset with a RMS error of 4.7% for a power fit and 3.4% for the logarithmic fit. The outlier, BWR8, is a different geometry than the other assemblies in the dataset and would therefore be expected to deviate from the main trend.

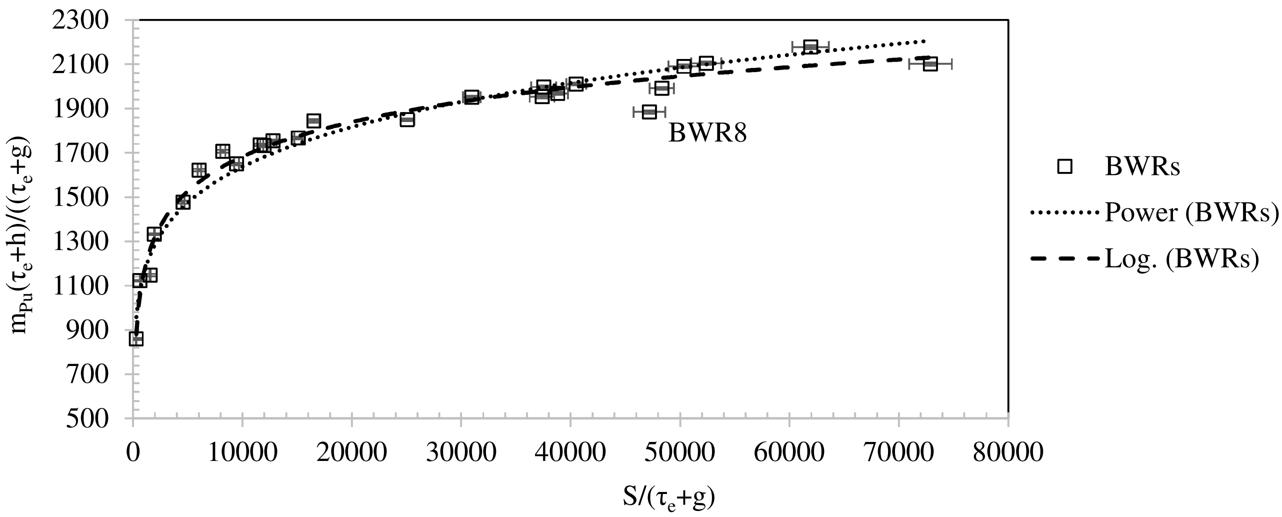

Figure 12. Power and logarithmic function relations between plutonium mass and singles rate, weighted by early die-away time and calibration constants for all BWR assemblies.

4.2. Burnup

The stochastic nature of production of spontaneous fission isotopes through the fuel irradiation process causes the total number of emitted neutrons from a spent fuel assembly to be related to the burnup of the assembly via a power function. By plotting the burnup values of the assemblies against the detected singles rates, a power function relationship can be observed as shown in Figure 13 and Figure 14, to find the RMS error in burnup prediction in the 25 PWR and 25 BWR assemblies. BU values in the BWR assemblies have been corrected for the location of the central measurement and are therefore labeled ABU for adjusted burnup. Using this power function correlation, burnup is determined with an RMS error of 7.5% for the PWRs and 6.8% for the BWRs.

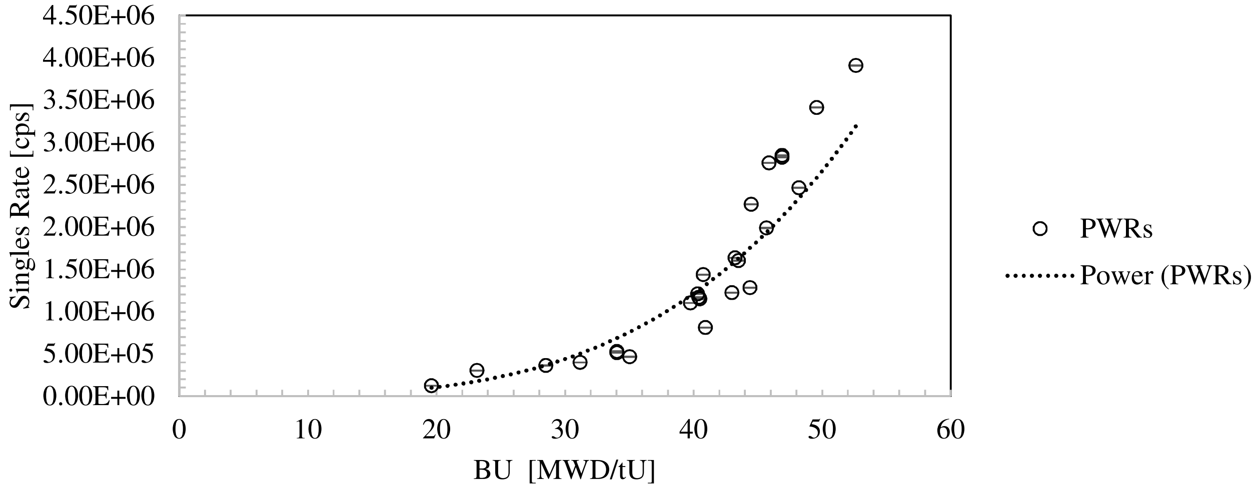

Figure 13. Burnup (BU) versus singles rate for 25 PWR assemblies with power function fit.

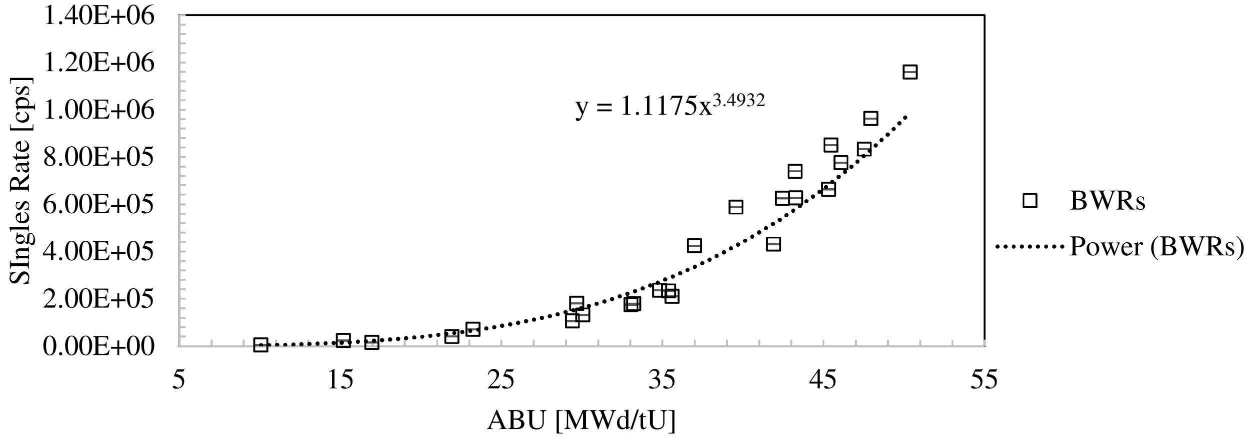

Figure 14. Adjusted burnup (ABU) versus singles rate for 25 PWR assemblies with power function fit.

4.3 Initial enrichment

The initial enrichment of an assembly not only determines the starting fissile mass but also how rapidly neutron absorbers grow in the system from variations in the ^{238}U mass. The relative amounts of neutron absorbers and fissile isotopes are what determine the multiplication of a spent fuel assembly. Therefore, because early die-away time is a measure of multiplication, the initial enrichment can be estimated using the predicted burnup value corrected for multiplication. Because initial enrichment affects multiplication via fissile mass more than burnup does via neutron absorbers, the impact of the burnup quantity on the function is reduced by raising it to the power of 0.5 or 0.25 for PWRs and BWRs respectively, optimized values for these datasets. The fitting parameters demonstrate that the burnup more strongly impacts the measured signal for PWRs than for BWRs. Early die-away time is multiplied by the reduced burnup and is used to determine initial enrichment. With this correlation, initial enrichment is determined with an RMS error of 6.3% for the PWRs and 7.0% for the BWRs. PWR24 is an outlier in the dataset as shown in Figure 15. This is because PWR24 has very low burnup and initial enrichment, and relatively long cooling time. The inherent signal is therefore extremely weak, resulting in a difficult-to-characterize assembly. If we exclude the PWR outlier (PWR 24), which is particularly apparent on the curve in Figure 15, then the RMS error for the PWRs drops to 3.9%.

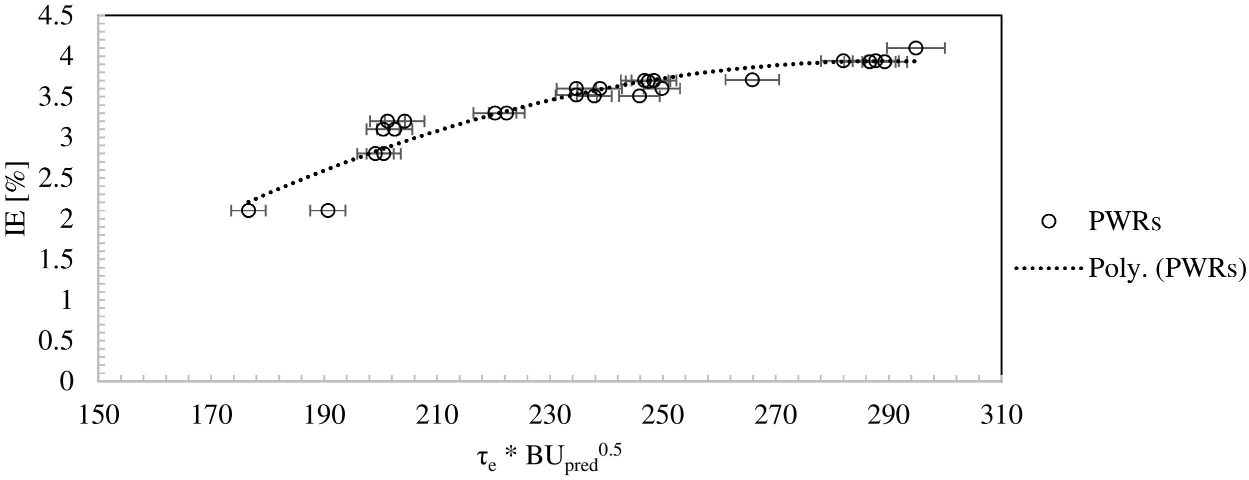

Figure 15. Initial enrichment (IE) versus early die-away time (τ_e) multiplied by adjusted predicted burnup ($BU_{pred}^{0.5}$), fit with a second-order polynomial, for PWRs.

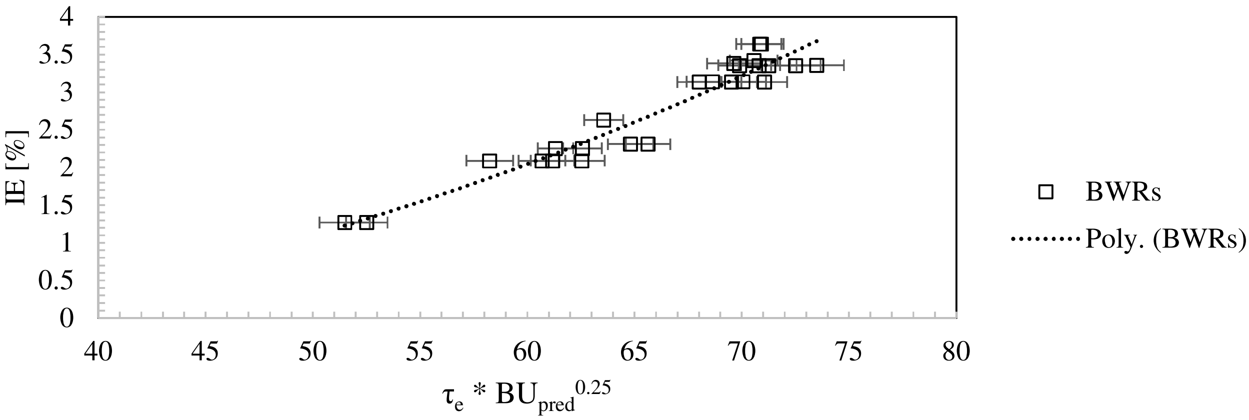

Figure 16. Initial enrichment (IE) versus early die-away time (τ_e) multiplied by adjusted predicted burnup ($BU_{pred}^{0.25}$), fit with a second-order polynomial, for BWRs.

4.4 Cooling time

As an assembly cools after it leaves a reactor, the gamma-emitting fission products decay in a predictable fashion, resulting in a slow decrease in the total gamma count rate. The burnup and initial enrichment of the assembly also factor into the detected gamma rate because the number of gammas emitted depends on how many fission products are produced, which is proportional to burnup. Therefore, we can fit a curve relating predicted burnup, early die-away time (which encompasses multiplication), and cooling time to the total gamma emissions measured in the ion chamber during the static center measurements to predict cooling time, as shown in Figure 17. Using this power fit, cooling time is determined in the PWR assemblies with an RMS error of 16.0% and the BWR assemblies with an RMS error of 14.0%. Note that gamma spectral analysis using an HPGe or LaBr detector in past measurements of the same assemblies is associated with smaller errors in predicting cooling time

[18]. In the future work we consider integrating spectral passive gamma and DDSI data to improve the CT estimations.

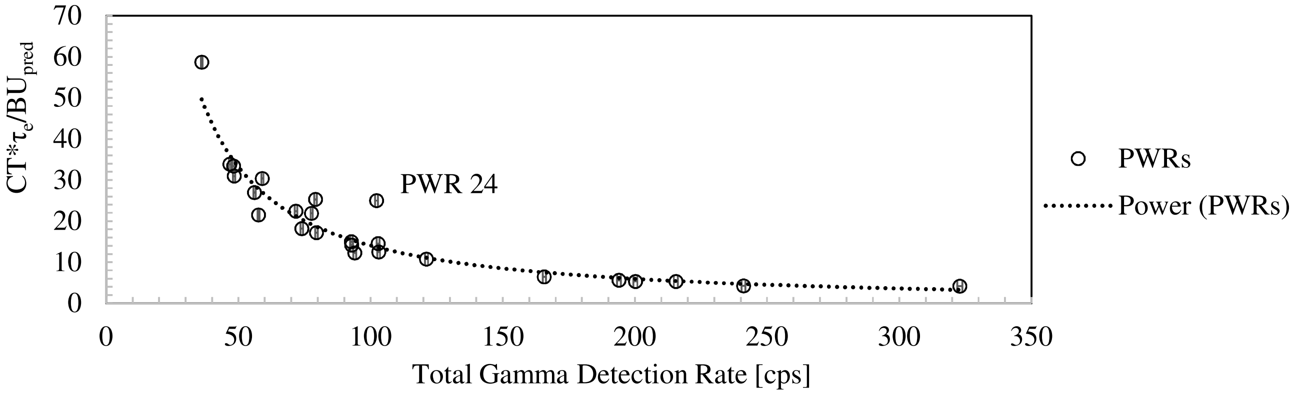

Figure 17. Relation between cooling time (CT), early die-away time (τ_e), and predicted burnup (BU_{pred}) to total gamma detection rate for PWRs.

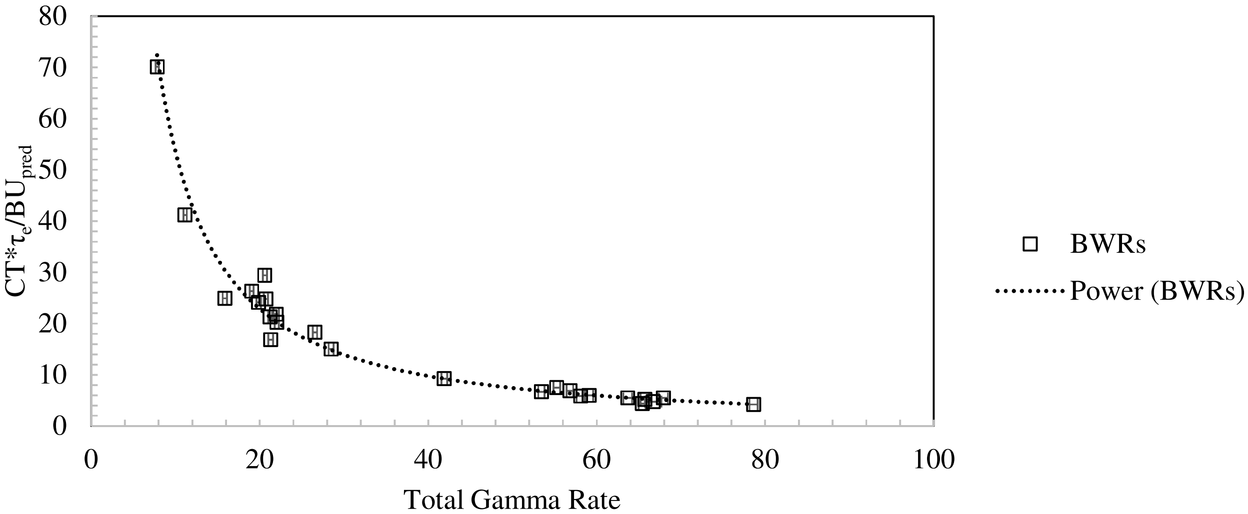

Figure 18. Relation between cooling time (CT), early die-away time (τ_e), and predicted burnup (BU_{pred}) to total gamma detection rate for BWRs.

This method of cooling time determination performs well for <20-year cooled assemblies, but it begins to break down for assemblies with higher cooling times. This holds true for both PWRs and BWRs. The primary passive neutron emitter in spent fuel, ^{244}Cm, has a half-life of 18.1 years, and as this isotope decays, the primary passive neutron source changes.

4.3. Fissile mass determination

Residual fissile mass in a spent fuel assembly is a function of initial enrichment and burnup. Multiplication is essentially a measure of balance between fissile mass and neutron absorbers, so it can also be included in an expression for determining fissile mass to improve the estimate. A method was developed using these three parameters to compute fissile mass in the PWR and BWR assemblies, as shown in Figure 19 and Figure 20, respectively.

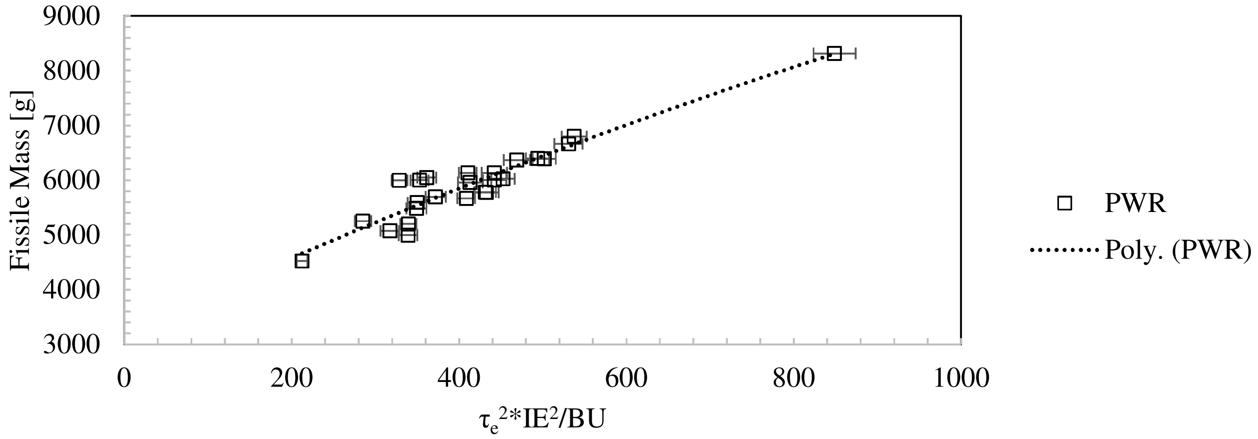

Figure 19. Fissile mass as a function of early die-away time, predicted initial enrichment, and predicted burnup for PWR assemblies.

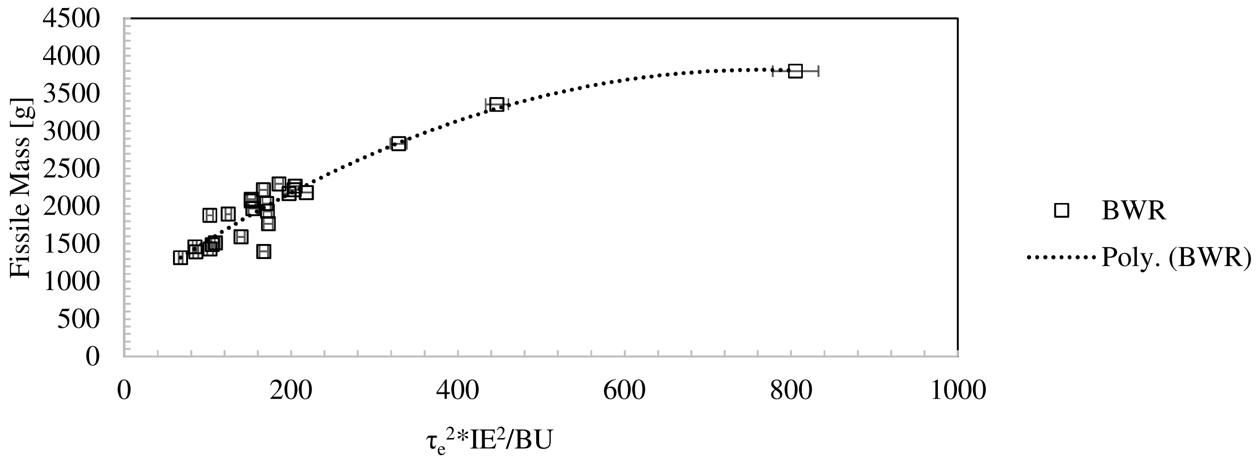

Figure 20. Fissile mass as a function of early die-away time, predicted initial enrichment, and predicted burnup for BWR assemblies.

Error bars on the x values were particularly large for this method that uses both predicted initial enrichment and burnup. The resulting fissile mass was determined for the data sets with RMSEs of 4.3% for the PWRs and 11.4% for the BWRs. As was the case with total Pu, the large axial variation of fissile mass and the single measurement point introduce systematic error into the fissile mass estimate. If we instead consider only the fissile mass in the region of fuel that is within the field of view of the neutron detectors, the estimate for the BWRs improves slightly to a RMSE of 10.9%.

5. Conclusions

A series of field trials measuring spent power reactor fuel with the DDSI system were successfully conducted at the Clab facility in Sweden. The field trial results represent the culmination of a 10 year effort to develop and deploy DDSI, a unique nondestructive assay tool and the only existing power reactor fuel measurement system capable of neutron coincidence counting with list-mode data collection. 50 unique PWR and BWR assemblies were measured, several of them in multiple locations, and each underwent an axial scan. Algorithms were developed to determine the following fuel assembly characteristics: multiplication, elemental plutonium mass, initial enrichment, burnup, cooling time, and fissile mass. Several of the algorithms were developed a priori in simulation space, and the methods were successfully demonstrated on the measured data as reported in this work. Additional algorithms, such as initial enrichment determination, were developed using the measured spent fuel data. Although not explicitly examined in this research, expected fissile or Pu mass can be compared to measured estimates, and deviations from declarations could be identified in order to meet the partial defect detection goal of this study. The results in this work demonstrate that the DDSI concept is capable of characterizing spent power reactor fuel with levels of accuracy that are compatible with the requirements and objectives of various applications such as safeguards verification or facility material control and accounting.

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