ANALYSIS OF PNAR SPENT FUEL SAFEGUARDS MEASUREMENTS USING THE ORIGEN DATA ANALYSIS MODULE

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ABSTRACT
This paper summarizes the analysis of the Passive Neutron Albedo Reactivity (PNAR) measurements using the ORIGEN data analysis module for 23 boiling water reactor spent fuel assemblies that were performed in Finland under an international collaboration on spent fuel safeguards verification methods. PNAR is part of the proposed integrated nondestructive system to be used for safeguards verifications at the planned Finnish encapsulation facility. Besides measuring passive neutron and gamma emission rates from an assembly like a Fork detector, PNAR also measures the PNAR ratio, which is expected to correlate with the fissile content in the assembly. The emission rates and PNAR ratio can be used to verify the operator declarations and the fissile content of an assembly, respectively. The analysis was performed using the ORIGEN Data Analysis Module, which was originally developed for predicting Fork detector neutron and gamma signals for spent fuel measurements and has been integrated into the Integrated Review and Analysis Package developed by Euratom and the IAEA. The Module includes the ORIGEN burnup analysis code and integrates detector response functions pre-generated using MCNP to predict detector signals in several seconds per assembly. In this study, new response functions specific to PNAR measurements were generated for ORIGEN Module. The study also analyzes impacts of using detailed fuel design and operation information vs. standard safeguards information on results calculated by ORIGEN Module. Using detailed information reduced the standard deviation of relative differences between calculated and measured neutron count rates among the 23 assemblies from ~10% to ~4%. The results obtained using standard safeguards information for these PNAR measurements were similar to those obtained for the Fork detector. A clear trend was found between the calculated net neutron multiplications and the measured PNAR ratios of the 23 assemblies. This paper describes how ORIGEN Module calculates the expected PNAR neutron and gamma signals and PNAR ratio and how they compare with corresponding measured values.

Keywords: spent fuel safeguards, PNAR, ORIGEN

1. INTRODUCTION
This paper summarizes the analysis of the Passive Neutron Albedo Reactivity (PNAR) measurements using the ORIGEN data analysis module for 23 boiling water reactor spent fuel assemblies that were performed in Finland under Action Sheet 65. This action sheet is an international collaboration among the US Department of Energy (DOE)/National Nuclear Security Administration (NNSA), represented by Los Alamos National Laboratory (LANL) and Oak Ridge National Laboratory (ORNL), the European Atomic Energy Community (Euratom), and the Radiation and Nuclear Safety Authority of Finland (STUK) to collaborate on spent fuel safeguards verification methods in context of the Finnish spent fuel encapsulation/repository system. PNAR is part of the proposed integrated nondestructive system to be used for safeguards verifications at the planned Finnish encapsulation facility [1] [2]. Like a Fork detector, PNAR measures passive neutron and gamma emission rates from a spent fuel assembly. PNAR also measures the PNAR ratio, which is a ratio of the neutron signal of the PNAR measurement without the Cd liner to that with the Cd liner in place. Tobin et. al. demonstrated using MCNP simulations that the PNAR ratio is directly correlated with the net neutron multiplication of an

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assembly [3], which is in turn dependent on the amount of fissile content and neutron absorbers present in an assembly. Therefore, the PNAR ratio provides a means to verify the fissile content in a spent fuel assembly.

Predictions of the PNAR neutron and gamma signals and the PNAR ratio of a particular spent fuel assembly based on the provided operator declarations (e.g., burnup, initial enrichment, cooling time) of the assembly will aid the safeguards inspectors to determine whether the measured PNAR signals and ratio are consistent with the assembly's declarations. A spent fuel assembly is a complex system that contains thousands of isotopes, whose quantities vary according to a number of factors such as the initial uranium loading, burnup, moderator density, and etc. of the assembly. Predictions of spent fuel assembly measurement signals are difficult (if not impossible) to make without the assistance of a sophisticated computer code. The ORIGEN data analysis module [4] (referred to as the ORIGEN Module in this paper) was previously developed to predict Fork detector spent fuel measurement signals in real time and has been integrated into the Integrated Review and Analysis Program (IRAP) developed by Euratom and the International Atomic Energy Agency. This module includes the ORIGEN burnup analysis code distributed as part of the SCALE (version 6.1.3) nuclear systems modeling and simulation suite [5] and integrates detector response functions pre-generated by using the Monte Carlo N-Particle (MCNP) code [6] to predict the detector signals in several seconds per assembly. The ORIGEN Module has been benchmarked against Fork measurement data for over 300 light water reactor assemblies [7], and it was recently expanded to VVER-440 assemblies for Fork detector measurements [8] in addition to BWR and pressurized water reactor (PWR) assemblies.

In this study, the ORIGEN Module was updated for the PNAR measurements by generating new response functions. The updated ORIGEN Module was then used to analyze PNAR measurements of the 23 BWR assemblies. Two datasets for the fuel design and operating conditions of these 23 assemblies were provided: (1) a set of basic data (e.g., assembly-average burnup, initial enrichments) similar to the operator declarations in a typical safeguards inspection (referred to as safeguards data in this report), which was provided by STUK and (2) a set of detailed data (e.g., detailed burnup and moderator density values along the height of the assemblies) provided by the reactor operator Teollisuuden Voima Oyj (TVO). Both sets of data were used in the calculations with the ORIGEN Module and the results are presented in this paper. The net neutron multiplication was also calculated with the ORIGEN Module and compared with the measured PNAR ratio. More comprehensive analysis of the PNAR measurement using the ORIGEN Module can be found in Reference [9].

2. PNAR MEASUREMENTS

The PNAR measurements were described in detail elsewhere [1] [2], so only a brief summary is provided here. The measurements were performed by STUK in July 2019 on 23 different BWR assemblies at the spent fuel interim storage facility at Olkiluoto Nuclear Power Plant. The cooling times of these assemblies ranged from 6.2 to 35.1 years, and burnups ranged from 18,589 to 49,698 MWd/tU. The assemblies included seven different assembly designs with three different assembly lattices (8×8, 9×9, and 10×10). Three assemblies experienced noncontinuous cycles in which they were removed from the core after irradiation, stored outside of the core for one or more cycles, and reinserted in the core for further irradiation. Measurements were made approximately 1.4 m from the bottom of the assembly. There are approximately 0.4 m of support structures and natural U at the bottom of the assembly below the enriched U zone [1]. The operator simulation data by TVO for these assemblies were provided for 25 equal-length axial nodes. Assuming that a natural U zone was present in the first bottom node (~15 cm) of most assemblies, the measurements were made ~115 cm above the bottom of the active fuel, which correspond to node 8 from the bottom of the assembly. The measured PNAR signals include the neutron and gamma count rates and the PNAR ratio from each assembly.

3. GENERATION OF PNAR RESPONSE FUNCTIONS

The response functions record the MCNP-simulated detector responses due to a source neutron or photon particle emitted from the spent fuel. 3D MCNP models of the measurement system are needed to adequately calculate the PNAR detector response caused by a neutron or photon originated in the spent fuel assembly, given the axial and radial variations of burnup values, isotopic compositions, and radiation emission sources in the spent fuel assembly. Only the axial burnup profiles were considered for response function generation in this study because the radial burnup variation is not nearly as important as the axial variations.

Two BWR assembly designs were selected for generating the spent fuel isotopic compositions and neutron/gamma source terms for use in the response function generation: (1) a SVEA64 design as representative for all 8×8 assemblies and (2) a SVEA100 design as representative for all 10×10 and 9×9 assemblies. Both representative BWR assemblies have a burnup of 35 GWd/tU, an initial enrichment of 3.0%, and a cooling time of 20 years. The 3D spent fuel isotopic compositions and neutron and gamma source terms, which are needed in the response function generation, of the two representative BWR assemblies were calculated by using the ORIGAMI code in SCALE (version 6.2.4) [10] based on a representative 25 node axial profiles of burnup and moderator density.

Figure 1 shows the 3D MCNP model used to generate the response functions for the PNAR measurement of a SVEA64 spent fuel assembly, which is consistent with the actual configuration of the PNAR instrument used in the measurement. The PNAR instrument is placed 115 cm above the bottom of active fuel. The top part of the assembly is not shown in the left figure due to limited space. The Cd liner is placed between the instrument and the fuel assembly in the "with Cd liner" case; the liner is moved 60 cm downward in the "without Cd liner" case, as labeled in this figure. There are four detector pods in PNAR, one on each side of the assembly. Each detector pod contains a ^{3}He tube for neutron detection and an LND 52110 ion chamber for gamma detection. More details of the PNAR instrument can be found in other works [3].

For the PNAR neutron response functions, 20 discrete source neutron energy bins were used to span 0.01–20 MeV with each energy bin simulated in a separate MCNP model. Since most neutrons are born at ~2 MeV in spent fuel, this neutron energy discretization is deemed sufficient. In each MCNP model, a fixed-source calculation was performed with MCNP6 (version 6.1) [6] with the neutron source particles sampled uniformly in the radial direction of the fuel assembly but nonuniformly in the axial direction based on the calculated neutron emission probability along the assembly axis. The neutron capture rates in the ^{3}He gas in all the PNAR ^{3}He tubes were tallied in these models to mimic the PNAR neutron count rates. The gamma count rates are calculated based on tallies for gamma dose rates deposited in the gas of the ion chamber, given that the gamma count rates are expected to be proportional to dose rates. ANSI/ANS 1977 flux-to-dose factors [6] are used to convert gamma flux into dose rates. Sufficient neutron and gamma particle history were used in these MCNP calculations and less than 0.5% statistical uncertainties were achieved in the neutron and gamma response functions at all energy bins except for the bin with the lowest energy.

Figure 2 illustrates the response functions for the PNAR neutron count rates for the modeled SVEA64 assembly design. Neutron response functions for the "without Cd liner" case are 4–24% higher than those for the "with Cd liner" case at varying energies, which is expected because the Cd liner absorbs thermal neutrons returning to the assembly from surrounding materials and thus reduces induced fissions in the assembly. Response functions generated for the SVEA100 design were similar to the ones shown here, but they are not presented here for brevity.

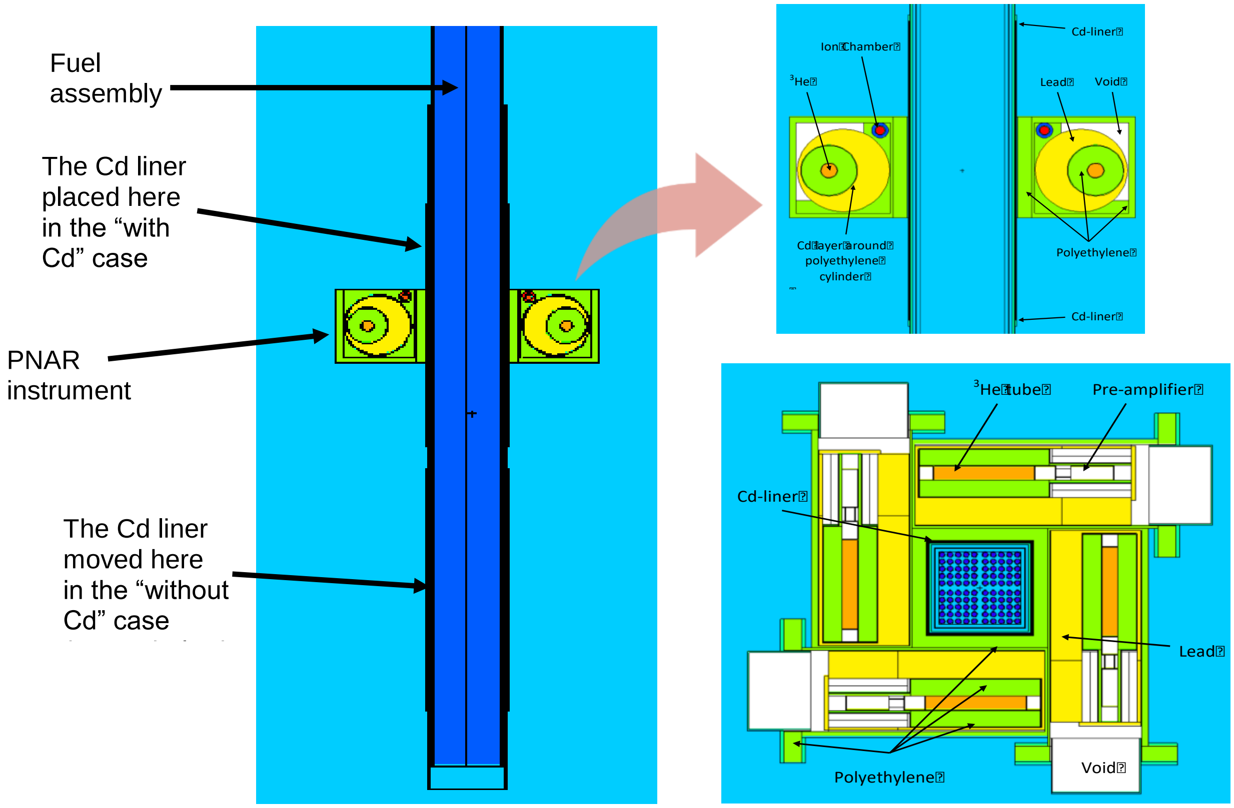


Figure 1. Side view of the MCNP model of the PNAR measurement with an assembly in the middle (top part of the assembly is not shown here) (left); magnified view of the PNAR detector (upper right); top view of the MCNP model of the PNAR measurement (lower right).

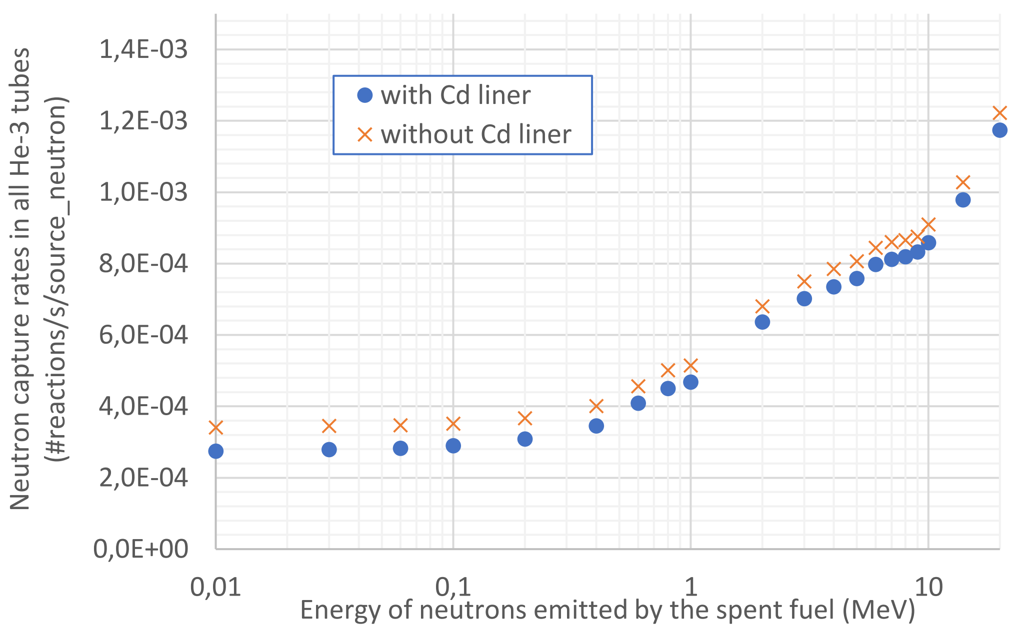


Figure 2. Response functions for the PNAR neutron count rates for the SVEA64 assembly design.

4. CALCULATION OF NET NEUTRON MULTIPLICATION

As mentioned in Section 1, the PNAR ratio is expected to be correlated with the net neutron multiplication (*Mult*). Therefore, it is important for this ORIGEN Module to calculate the *Mult* of a given spent fuel assembly. To determine the *Mult* for each assembly, the following equation was used:

$$Mult = 1/(1 - k_{eff}) \simeq 1/(1 - k_{\infty}\,(1 - L)), \tag{1}$$

where k_{eff} and k_{∞} are the effective and infinite neutron multiplication factor, respectively, of the assembly, and L is the neutron leakage factor. ORIGEN calculates k_{∞} as the ratio of total neutron production (fission cross section multiplied by the number of neutrons per fission) over total neutron absorption. There is no geometry associated with this parameter other than the weighting of the neutron cross sections for the assembly design. L is calculated as $L = 1- k_{eff} / k_{\infty}$, where k_{eff} is determined by MCNP for a given fuel assembly geometry and composition. The leakage factor is largely constant for a fixed PNAR measurement configuration and is only weakly dependent on the fuel composition. L was calculated to be 0.55 and 0.56 for the 8 × 8 and 10 × 10 SVEA assembly designs, respectively. For the remaining three 9 × 9 assemblies, L is assumed to be 0.56. The values stored in the detector response files are the neutron non-leakage probabilities $(1 - L)$.

5. CALCULATION RESULTS AND COMPARISON WITH MEASUREMENTS

Calculations of the PNAR neutron and gamma signals were performed with the ORIGEN Module by using both safeguards data and operator data as inputs in the calculations. The results for the PNAR measurements analyses obtained with each of these two datasets are presented in this section. The safeguards data provided by STUK included assembly type, assembly-average burnup, assembly-average initial enrichment, cooling time, and U and Pu mass at discharge from the reactor for each of the 23 assemblies [9]. The initial U mass in each assembly was inferred by using the ORIGEN burnup code based on the provided information for these assemblies.

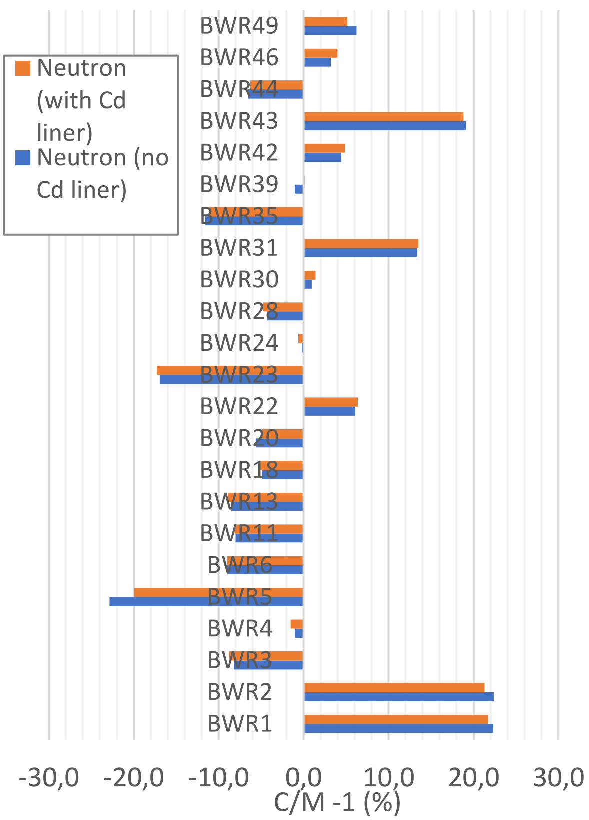
Besides the initial U loading, initial enrichment, and cooling time for each of the 23 assemblies, the operator data also includes node-by-node burnup and moderator densities along the height of each assembly, which were extracted from the CASMO/SIMULATE SNF output files provided by the reactor operator, TVO. Burnup values and moderator densities of node 8 from the operator data were used in the ORIGEN calculations to reflect the assembly characteristics in proximity to the axial measurement position of the assembly. The main difference between using safeguards data vs. operator data in the ORIGEN Module calculations in this work is that the former uses assembly-average burnup and moderator density (0.4555 g/ cm^3) values, whereas the latter uses nodal burnup and moderator density values near the measurement height. In both cases, the ORIGEN Module simulations assumed a continuous cycle history and a constant specific irradiation power of 24 MW/MTU, which is a representative power level for BWR assemblies. Such simplification was necessary because the detailed cycle history is usually not included in safeguards data.

Once the raw PNAR neutron and gamma count rates were calculated by the ORIGEN Module, the neutron count rates are first multiplied by *Mult* to account for the induced fissions in each assembly and the gamma count rates were first adjusted to account for nonlinear response to gamma dose rate, which was previously found with LND 52110 ion chambers [7]. After that, the averages of the calculated and measured neutron and gamma count rates among the set of 23 BWR fuel assemblies were compared, and the ratios of the measured average signals to the calculated averages were taken and they are referred to as *scaling factors*. One scaling factor common for all 23 assemblies was used to scale the calculated neutron count rates of each assembly and another common scaling factor was used to scale the calculated gamma count rates for all 23 assemblies. The purpose of using these two scaling factors is to account for factors (e.g., electronic efficiency) that were not accounted for in the ORIGEN Module calculations. Such scaling factors can be replaced by calibration factors if sufficient PNAR measurement data have been collected by the same PNAR instrument at the same spent fuel pool.

Figure 3 (left) shows the relative differences between the measured PNAR neutron count rates and the calculated values from the ORIGEN Module by using safeguards data for each of the 23 assemblies. Figure 3 (right) shows a similar comparison but with the operator data used in the calculations. As shown in Figure 3 (left), the standard deviation among the 23 assemblies is 11.7% for the neutron count rates without the Cd liner and 11.3% for the rates with the Cd liner. Sixteen of the 23 assemblies have calculated neutron count rates within 10% of the measurement, and the other seven assemblies (BWR1, 2, 5, 23, 31, 35, and 43) have calculation-to-measurement relative differences between -22.9 and 22.4%. BWR1 and BWR2 are two of the three assemblies with noncontinuous cycle histories. These results are similar to the results seen in earlier studies with the Fork detector when typical safeguards data were used in the ORIGEN Module calculations for BWR assemblies [7], mainly due to BWR assemblies' complexities in the fuel design and operating conditions. As shown in Figure 3 (right), the calculated results from using operator data were significantly improved when compared with measurements than those from using safeguards data. The standard deviation among the 23 assemblies is 3.9 and 4.4% for the neutron count rates with and without the Cd liner, respectively. All assemblies' calculated neutron count rates are within 10% of the measurements, with 18 assemblies showing differences in the calculation-to-measurement of less than 5%.

Figure 4 (left) shows the relative differences between the measured PNAR gamma count rates and the calculated values from the ORIGEN Module by using safeguards data for each of the 23 assemblies. Figure 4 (right) shows a similar comparison but with the operator data used in the calculations. As shown in Figure 4 (left), the standard deviation among the 23 assemblies is 11.8 and 11.9% for the cases with and without the Cd liner, respectively. These differences would have been 16.3 and 15.4%, respectively, if adjustments were not made to account for the ion chamber nonlinear response. All assemblies, except for BWR1 and BWR31, had calculated gamma count rates within 16% of the measured values. As shown in Figure 4 (right), the standard deviation among the 23 assemblies is 10.9 and 11.0% for the cases with and without the Cd liner, respectively. All assemblies—except for BWR1, BWR13, and BWR30—show calculated rates that are within 15% from the measured values. The gamma count rate results are only marginally improved when operator data were used compared with the cases using safeguards data, which is unsurprising because the gamma emissions are less sensitive to nodal burnup and moderator density conditions than neutron emissions. The gamma results are also similar to those observed in the ORIGEN Module analysis of Fork detector measurements for BWR assemblies [7].

Figure 5 compares the measured PNAR ratio with the calculated *Mult* of each assembly with the safeguards data used in the calculations. The linear-fit trendline equation and the goodness of fit parameter (R^2) is also shown. The error bars (horizontal direction) on the measured PNAR ratios include the counting statistical uncertainty of the measurement and a repeatability uncertainty of 0.0013 [1]. The error bars for calculations are estimated as a sum of 2.7% uncertainty due to the variability in the assembly-to-assembly burnup profile at the measurement position and enrichment variability at the measurement position to that of the assembly-average, and a 2% uncertainty associated with the calculation of *Mult* [9]. No uncertainty has been assigned for cooling time or U mass. A detailed uncertainty analysis can be found in Reference [9]. As shown, the calculated *Mult* generally follows a linear trend with the measured PNAR ratio, confirming that there exists an underlying correlation between the two quantities, as a previous study suggested [3]. Most data points fall in a narrow range along the trendline that is overlapped with the uncertainty band. BWR1 was irradiated in non-continuous cycles and was also an initial core assembly. In previous studies that used the Fork detector [7], first cycle assemblies exhibited a systematic bias compared with other assemblies, likely due to unusual reactor operations of the start-up core. These reasons might explain why BWR1 deviated the most from the trendline. BWR5 has the lowest ratio of burnup to initial enrichment values based on both safeguards and operator data, indicating that it is the least "burned" assembly, and thus BWR5 is expected to have the highest ratio of fissile content to fission product absorbers remaining in the assembly. This would explain why BWR5 had the highest *Mult* and PNAR ratio. On the contrary, BWR49 has the highest ratio of burnup to initial enrichment values, indicating it is the most "burned" assembly, which explains why BWR49 had the lowest Mult and PNAR ratio. Similar trend was found between measured PNAR ratio and calculated *Mult* with operator data used in the calculations, and the results can be found in Reference [9].



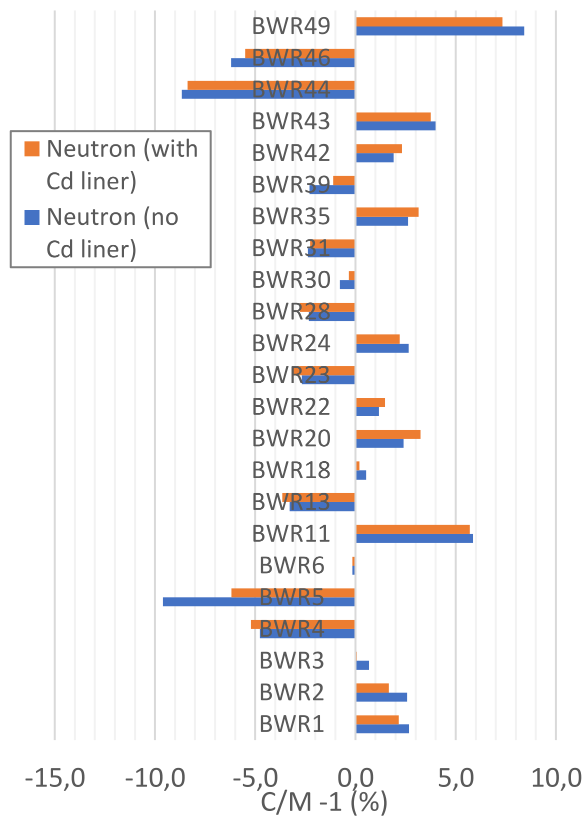


Figure 3. The neutron count rate relative differences between PNAR measurement (M) and calculation (C) using (left) safeguards data and (right) operator data for each assembly.

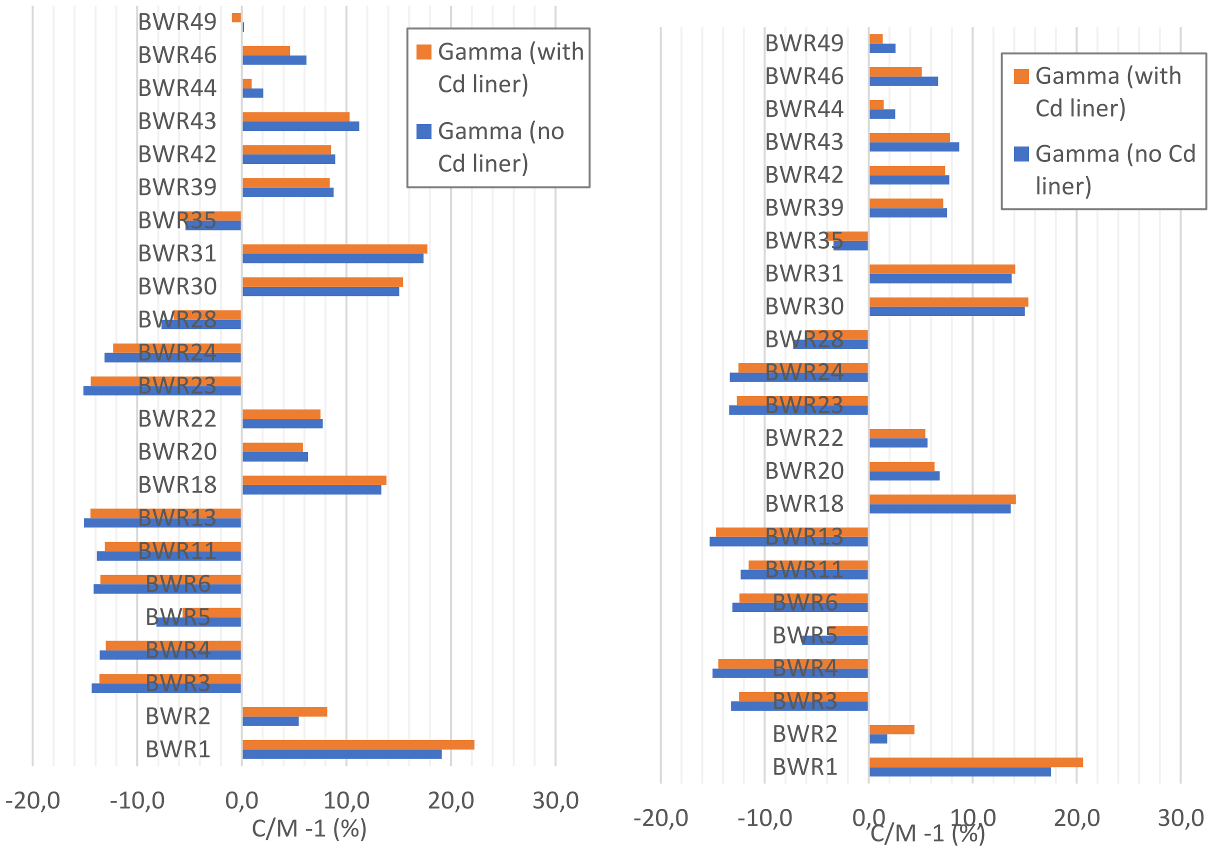


Figure 4. The gamma count rate relative differences between PNAR measurement (M) and calculation (C) using (left) safeguards data and (right) operator data for each assembly.

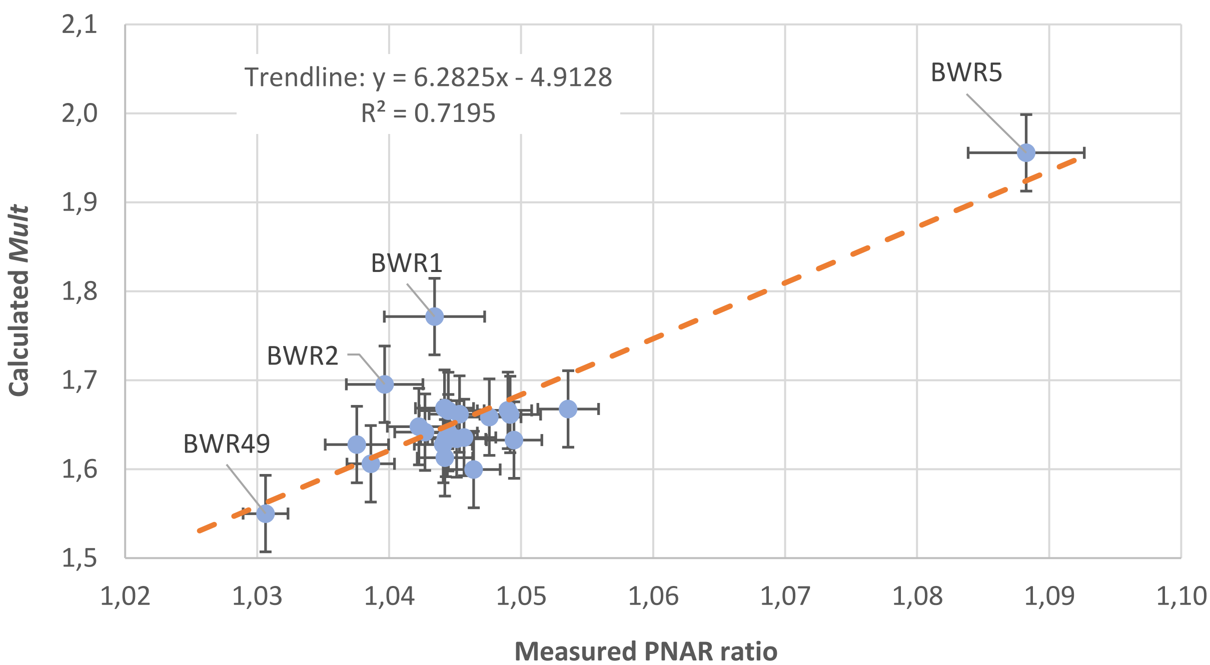


Figure 5. The calculated net neutron multiplication (*Mult*) using safeguards data vs. measured PNAR ratio.

6. SUMMARY AND CONCLUSIONS

The PNAR instrument is designed to measure the neutron multiplication of a fuel assembly to ensure that it is multiplying at a level consistent with the operator declaration and the expected fissile content in the assembly. The ORIGEN Module calculation results show that when typical safeguards data (typically including only assembly-average attributes) were used, the standard deviations between measured and calculated neutron and gamma detector count rates were similar to the results seen in earlier studies that used the Fork detector due to complexities in BWR assemblies. The agreement between the calculated count rates and the measurement values was improved by the added nodal burnup and moderator density values included in the operator data, which resulted in a standard deviation of the calculated-to-measured value among the 23-assembly set of ~4% for neutron and 10% for gamma count rates.

The measured PNAR ratio was analyzed by trending the PNAR ratio with the calculated net neutron multiplication (*Mult*) of the assembly since a correlation between them was expected but the exact correlation has not yet been established prior to this work. The results show a clear linear trend between the measured PNAR ratio and calculated *Mult*. Most data points fall in a narrow range along the trendline that is overlapped with the uncertainty band. The predicted assemblies with the highest and lowest *Mult* by the ORIGEN Module were same as the assemblies with the highest and lowest measured PNAR ratios.

In summary, based on comparisons between calculation and measurement, the results show that the ORIGEN Module can reasonably predict the PNAR neutron and gamma signals and *Mult*, which was found to follow a linear trend with the measured PNAR ratio. Because an ORIGEN Module calculation takes only seconds to complete, such predictions will be useful for the safeguards inspectors to draw conclusions in real time. Future work is recommended to investigate how to further reduce discrepancies between the calculated and the measured quantities while considering the complexities in fuel design parameter and operator conditions (e.g., enrichment zoning, part length rods, and noncontinuous cycles) that often exist in BWR assemblies.

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