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# Validation of ORIGEN for VVER-440 Spent Fuel with Application to Fork Detector Safeguards Measurements

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***Abstract:***

*The US Department of Energy National Nuclear Security Administration and the European Atomic Energy Community (Euratom) are collaborating with the Radiation and Nuclear Safety Authority in Finland to assess spent fuel verification methods for potential application to measurements at the planned spent fuel encapsulation plant and geological repository prior to final disposal in Finland. The fork detector (FDET) used to measure the neutron and gamma radiation from a given spent fuel assembly is an existing technology widely used by the International Atomic Energy Agency (IAEA) and Euratom for the safeguards verification of spent fuel operator declaration. Recently, an FDET data analysis software, referred to as the ORIGEN module, was developed and incorporated into the IAEA/Euratom Integrated Review and Analysis Program. This module uses the ORIGEN burnup code to calculate the nuclide concentrations and the neutron and gamma source terms in the spent fuel based on operator declarations. Then, to predict the expected FDET signals for a given fuel assembly, the module combines the source terms calculated by ORIGEN with FDET response functions that were pre-generated using MCNP models. The inspector can compare the calculated signals to the measured values in real time to identify possible anomalies in fuel assembly's operator declaration, integrity, or measurement. In this work, the capability of the ORIGEN module was extended from application to typical light water reactor fuels to use for VVER-440 assembly types. The accuracy of ORIGEN for this type of fuel assembly calculations was assessed by comparing calculated nuclide concentrations against destructive assay measurements for VVER-440 spent fuel. The performance of the ORIGEN module for FDET safeguards verification in routine inspections was assessed using FDET measurement data for 13 spent VVER-440 assemblies that were measured at the Loviisa Nuclear Power Plant in Finland, by comparing the calculated signals to the measured quantities. The results show that the calculated FDET neutron and gamma detector signals are generally within 12% of the measurements except for one assembly. These results are applicable to future safeguards verification in the planned Finland encapsulation plant, such as for passive neutron albedo reactivity safeguards measurements.*

**Keywords:** spent fuel safeguards; Fork; FDET; ORIGEN; safeguards verification; encapsulation

## 1. Introduction

Finland is anticipated to be one of the first countries in the world to open a geological repository for permanent storage of spent nuclear fuel [1]. The US Department of Energy National Nuclear Security Administration and the European Atomic Energy Community (Euratom) are collaborating with the Radiation and Nuclear Safety Authority (STUK) in Finland on spent fuel safeguards verification methods that can be implemented prior to the fuel's encapsulation and geological disposal in Olkiluoto, Finland. STUK is working with the International Atomic Energy Agency (IAEA) and Euratom to finalize the spent fuel safeguards verification requirements and approaches for the planned encapsulation plant and repository [1]. The approaches to nondestructive assay (NDA) measurements for safeguards have evolved over time. Because there will be no spent fuel storage pools in the encapsulation plant, the plan is to perform passive gamma emission tomography (PGET) [2] and passive neutron albedo reactivity (PNAR) [3] safeguards measurements under water in the reactor site storage pools for safeguards verification, before transferring the fuel to the encapsulation plant. As one of the primary NDA instruments for spent fuel safeguards, the fork detector (FDET) [4] may be used for special inspections, for example, to re-establish continuity of knowledge of an assembly. The two options under consideration are (1) using PGET + PNAR and (2) using PGET + FDET. PGET measures gamma rays emitted from a given assembly in multiple angular positions around the vertical axis using directionally collimated gamma detectors, in order to reconstruct a cross-sectional image of the assembly to

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verify the presence of the rods in the assembly. FDET measures the total neutron and gamma emissions from an assembly [4]. The IAEA and Euratom have used FDET routinely for spent fuel safeguards verification globally since the 1990s. PNAR is similar to FDET in that it also measures total neutron and gamma emissions, but it can also quantify the assembly's neutron multiplication—an indicator of fissile content—using measurements with and without a cadmium liner neutron absorber around the assembly. The combined measurements of PGET + FDET or PGET + PNAR can be used to verify both the integrity of a given fuel assembly and the operator declarations—two primary goals for spent fuel safeguards.

Given the complexities of tracking nuclide transmutations during irradiation and decay and predicting neutron and gamma source terms in a spent fuel assembly, simulations using sophisticated computer codes are generally needed to accurately predict the NDA signals from a spent fuel assembly measurement for safeguards verification. Oak Ridge National Laboratory (ORNL) and Euratom have collaborated to develop a data analysis module for FDET, to fully automate the prediction of neutron and gamma detector signals using operator declarations of a spent fuel assembly [4] [5], with a specialized module resulting from this effort. Such predictions can help the safeguards inspector to draw conclusions in real time on whether the operator declarations of a given fuel assembly are consistent with the measured signals. The new module, referred to as *the ORIGEN module* in this paper, is based on the ORIGEN isotope generation and decay code [6] and incorporates detector response functions developed using MCNP [7] models of the assembly and detector configuration. The ORIGEN module has been incorporated into the Integrated Review and Analysis Program (IRAP) software that is jointly developed and maintained by IAEA and Euratom. The ORIGEN module has been extensively benchmarked for typical pressurized water reactor (PWR) and boiling water reactor (BWR) assembly types [4] [5], but has not been benchmarked for VVER-440 assembly types, as FDET measurement data on those types of assemblies are scarce. In this work, the ORIGEN module's application was extended to VVER-440 assembly types by generating new ORIGEN reactor cross-section libraries, referred to as *ORIGEN libraries* in this paper, and new FDET response functions to support potential applications to the Loviisa reactors in Finland. This paper establishes ORIGEN's accuracy in predicting the fissile and radiation-emitting nuclides in VVER-440 fuel and applies the ORIGEN module to evaluate FDET measurements of 13 spent VVER-440 assemblies at the Loviisa Nuclear Power Plant.

## 2. VVER-440 assembly designs

VVER-440 fuel assemblies are used in two reactors in Finland and in several eastern European countries. The VVER-440 assembly has 126 fuel rods arranged in a hexagonal lattice, with one central tube that does not contain fuel. There are two basic types of VVER-440 assemblies: standard assemblies and control assemblies with fuel followers. The top section of such control assemblies contain neutron absorbing materials, and the bottom section contains fuel. These control assemblies move vertically in the core, with the follower section of the assembly displacing the control section as the assembly is moved upward in the core. The fuel followers have a slightly shorter active fuel length of 2,320 mm compared to 2,420 mm for standard assemblies, and therefore they contain less uranium. Since FDET measurements are usually performed near the center of the assembly—away from the ends and away from the control element—control assemblies are treated as standard assemblies for the purpose of burnup analysis.

The TVEL Fuel Company (TVEL), a Russian fuel supplier, manufactured assemblies for the Loviisa reactors with initial enrichments of 1.6, 2.4, and 3.6 wt% $^{235}$U [8] and a uniform enrichment distribution (i.e., all fuel rods in an assembly have the same enrichment). These enrichments are widely used in other VVER-440 plants. The general design characteristics of the TVEL VVER-440 assembly design are listed in Table 1.

| Description | Parameter value |
|---|---|
| Lattice pitch type | Triangular |
| Number of fuel rods | 126 |
| Number of non-fuel rods | 1 |
| Inner fuel pellet radius (cm) | 0.075 |
| Fuel pellet radius (cm) | 0.3783 |
| Cladding inner radius (cm) | 0.388 |
| Cladding outer radius (cm) | 0.455 |
| Fuel rod pitch (cm) | 1.22 |
| Fuel assembly pitch (cm) | 14.70 |

**Table 1:** VVER-440 TVEL assembly design data [8].

At higher enrichments, the designs can use either uniform enrichment or enrichment profiling, whereby the fuel rods may have different enrichments, with lower enrichment rods generally located near the periphery of the assembly. These higher enrichment assemblies may have some fuel rods that contain burnable absorbers such as gadolinium. Prior to 2009 Loviisa Nuclear Power Plant operated with a first-generation TVEL design for standard assemblies, with 4.0 wt% $^{235}$U enrichments without profiling or gadolinium absorbers, and with a second-generation fuel follower design (see Section 3 for details). Since 2009, the Loviisa has operated with a second-generation TVEL design with a radially profiled enrichment and six gadolinium absorber rods [8] in the fuel assembly. Fuel assemblies manufactured by British Nuclear Fuels Limited (BNFL)/Westinghouse have also been operated in Loviisa, with uniform enrichments of 3.7 and 3.8% without profiling [8]. While the

Westinghouse assembly design is similar to that of TVEL, the fuel rod diameter is slightly smaller, and the fuel rod pellets do not have a central hole to accommodate swelling and fission product gases.

## 3. Generation of origen libraries for Loviisa assembly designs

ORIGEN libraries are required for standalone ORIGEN calculations to predict the nuclide concentrations and neutron and gamma source terms in nuclear fuel following its irradiation and decay. These ORIGEN libraries can be pregenerated using the TRITON depletion sequence in SCALE [9] to create one-group neutron cross sections for the fuel as functions of assembly design, moderator density, fuel enrichment, and fuel burnup. A large array of libraries for various fuel assembly types is included in the standard SCALE code package [9]. ORIGEN libraries for some of the VVER TVEL designs used at Loviisa are available in the SCALE distribution, but several other VVER designs used at Loviisa are not available, including BNFL/Westinghouse designs and TVEL designs with 4.0 and 4.37% enrichments, as shown in Table 2.

New ORIGEN libraries were generated in this work for all of the VVER-440 assembly types shown in Table 2. ORIGEN libraries for the VVER-440 assembly types existing in SCALE were regenerated to maintain consistency, by using the same set of nuclear data to prevent potential biases caused by different sets of nuclear data. The most recent version of the SCALE code package (version 6.2.3) [9] was used. The ORIGEN libraries were generated with the TRITON depletion sequence in SCALE with two-dimensional (2D) models of the fuel assemblies. TRITON iteratively couples the 2D neutron transport code NEWT with ORIGEN [9]. Under TRITON, the neutron spectrum calculated by NEWT is used to collapse the multigroup neutron cross sections into one-group cross sections, which ORIGEN uses to calculate reaction rates and the time-dependent evolution of nuclide compositions during irradiation and decay. The SCALE 252-group neutron cross section library, used in these TRITON calculations, contains cross sections for more than 400 nuclides, most of which are from ENDF/B-VII.1 [10] with the remainders from JEFF-3.1/A activation libraries [11]. The collapsed one-group cross sections that are calculated when running TRITON are automatically saved as ORIGEN library files for future standalone burnup-dependent ORIGEN calculations. Standalone ORIGEN calculations take only a small fraction of the computation time required for a TRITON calculation.

Figure 1 shows the TRITON models developed in this work for three different VVER-440 assembly designs used in Finland: (a) the TVEL design with flat enrichment profile (first generation); (b) the TVEL design with profiled enrichment (second generation); and (c) the BNFL/Westinghouse NOVA-E3 design with flat enrichment. In Figure 1 (b), the corner rods have lower enrichments and the green colored rods contain gadolinium as a burnable absorber. The central holes in each rod of the TVEL designs are shown in this figure.

| Designs | Enrichment(s) (wt% $^{235}$U) | Profile |
|---|---|---|
| **Assembly designs with libraries available in SCALE** | | |
| TVEL | 1.6, 2.4, 3.6 | Flat |
| TVEL | 3.82 (avg.*) | Profiled |
| TVEL | 4.25 (avg.) | Profiled |
| TVEL | 4.38 (avg.) | Profiled |
| Assembly designs with libraries not available in SCALE but used at Loviisa Nuclear Power Plant | | |
| TVEL standard | 4.0 | Flat |
| TVEL Gen-II | 4.0 | Flat |
| TVEL Gen-II | 4.37 (avg.) | Profiled |
| BNFL/Westinghouse NOVA E-3 | 3.7, 3.8 | Flat |

*average of radially varying enrichment across an assembly

**Table 2:** VVER-440 assembly designs for ORIGEN libraries [8] [9].

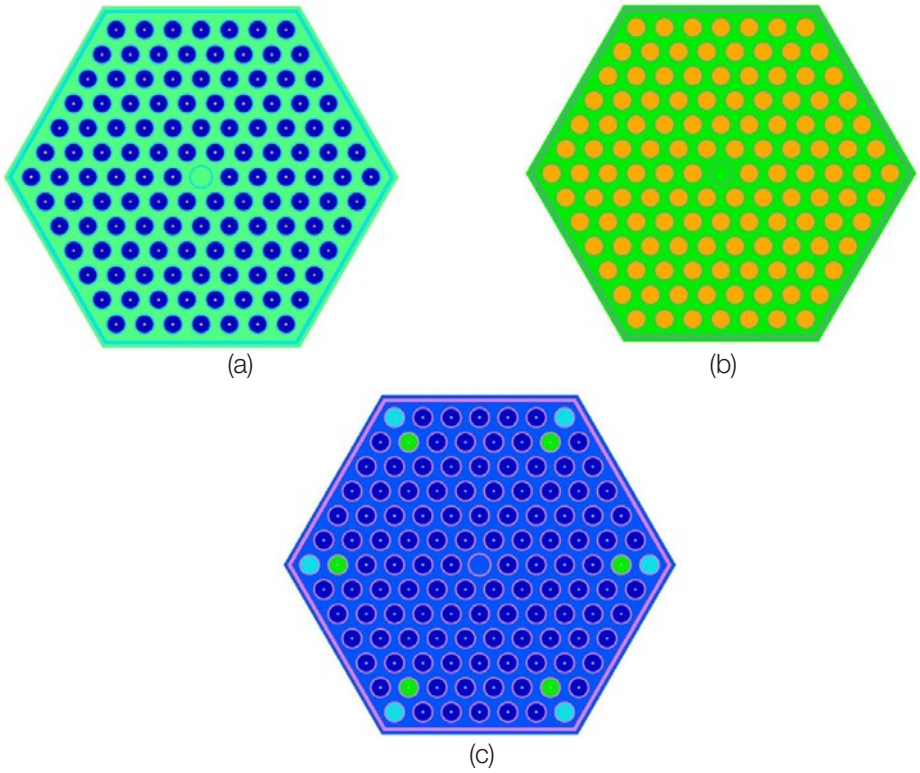


**Figure 1:** TRITON models of VVER-440 assembly designs: (a) TVEL design with flat enrichment; (b) the TVEL design with profiled enrichment **[8]**; (c) BNFL/Westinghouse NOVA-E3 design with flat enrichment.

## 4. Experimental data on nuclide concentrations

It is important to validate the new VVER-440 ORIGEN libraries with measurement data before using them for nuclear safety and safeguards applications. Since the NDA safeguards measurements of spent fuel are mostly performed on individual fuel assemblies, and the ORIGEN calculations using the ORIGEN libraries are designed for assembly-average nuclide concentrations, the ideal measurement data on nuclide concentrations for validation would be on an assembly level as well, e.g., an individually reprocessed assembly; however, such data are not available in the public domain for VVER-440 assemblies.

Measurements of nuclide concentrations in VVER-440 spent fuel samples reported in literature that were performed using destructive assay (DA) in Russia are used as a benchmark dataset in this work. The samples were usually small axial segments (~1 cm long) cut from spent fuel rods, then dissolved in acid solutions and prepared for radiochemical analysis. The concentrations of important nuclides were measured using various techniques, including mass spectrometry and gamma spectroscopy [12]. Many of these measurements were compiled into the international database of spent fuel compositions [13] hosted and maintained by the Organisation of Economic Cooperation and Development/Nuclear Energy Agency [14]. This database, called the Spent Fuel Isotopic Composition (SFCOMPO), was compiled with the contributions and cooperation of many countries. Early efforts to compile and upload VVER-440 and VVER-1000 data [15] included DA data from three VVER-440 reactors operated in Russia: Kola-3, Novovoronezh-3 (Novo-3), and Novovoronezh-4 (Novo-4). A total of 47 spent fuel samples were measured from 5 different fuel assemblies. The samples cover initial enrichment values ranging from 3.3 to 4.38% $^{235}$U. Measurements for all samples include uranium and plutonium isotopes and $^{244}$Cm, a major neutron-emitting source. Measurements of $^{137}$Cs and $^{154}$Eu, both major gamma emitters and frequently used as burnup indicator nuclides, were available for 20 samples (Kola-3 samples).

A summary of the VVER-440 measurement data is provided in Table 3, along with the main fuel characteristics [16] [17] [18]. The locations of the measured fuel rods in the assembly were determined from the fuel rod identification as shown in Table 3, and the schematic of the assembly layout is shown in Figure 2. The rod locations can be important since the rods adjacent to the central instrument tube and at the assembly periphery could be subject to more moderated neutronic environment compared to other rods, which would affect the nuclide concentrations. Larger deviations might be expected for these peripherical samples since they are less representative of the assembly average. Samples located near the periphery or instrument tube are highlighted in Table 3. The rod locations are color coded in this figure, with each color representing samples from the same reactor, and the same color coding is used in the figures in later of this section. Positions 25, 63, and 69 were used by samples from two reactors and therefore they were double colored.

| Sample[a] # | Reactor | Assembly | Fig. 2 fuel rod ID | Sample ID | Sample burnup (GWd/tU)[b] | Sample enrichment (wt% $^{235}$U) | Cooling time[c] (year) | Axial Elevation[d] (mm) |
|---|---|---|---|---|---|---|---|---|
| 1 | Kola-3 | 144-46879 | 96 | 5 | 35 | 4.38 | 7.3 | 66 |
| 2 | Kola-3 | 144-46879 | 61 | 13 | 47.9 | 4.37 | 7.3 | 185 |
| 3 | Kola-3 | 144-46879 | 61 | 53 | 62.7 | 4.37 | 7.3 | 825 |
| 4 | Kola-3 | 144-46879 | 96 | 63 | 51.2 | 4.38 | 7.3 | 196 |
| 5 | Kola-3 | 144-46879 | 61 | 76 | 42.3 | 4.37 | 7.3 | 108 |
| 6 | Kola-3 | 144-46879 | 62 | 81 | 31.8 | 4.38 | 7.3 | 44 |
| 7 | Kola-3 | 144-46879 | 96 | 98 | 58.3 | 4.38 | 7.3 | 355 |
| 8 | Kola-3 | 144-46879 | 61 | 124 | 32.7 | 4.37 | 7.3 | 43 |
| 9 | Kola-3 | 144-46879 | 62 | 165 | 40.8 | 4.38 | 7.3 | 107 |
| 10 | Kola-3 | 144-46879 | 96 | 169 | 40.8 | 4.38 | 7.3 | 109 |
| 11 | Kola-3 | 144-46879 | 61 | 189 | 57.6 | 4.37 | 7.3 | 314 |
| 12 | Kola-3 | 144-46879 | 96 | 718 | 61.4 | 4.38 | 7.3 | 549 |
| 13 | Novo-3[e] | RP-3371A | 63 | 1 | 17.1 | 3.3 | 3.3 | 1875 |
| 14 | Novo-3 | RP-3371A | 97 | 2 | 14.2 | 3.3 | 3.3 | 2170 |
| 15 | Novo-3 | RP-3371A | 97 | 3 | 8.7 | 3.3 | 3.3 | 200 |
| 16 | Novo-3 | RP-3371A | 58 | 4 | 13.4 | 3.3 | 3.3 | 200 |
| 17 | Novo-3 | RP-3371A | 63 | 5 | 13.9 | 3.3 | 3.3 | 375 |
| 18 | Novo-3 | RP-3371A | 97 | 6 | 11.7 | 3.3 | 3.3 | 300 |
| 19 | Novo-3 | RP-3371A | 97 | 7 | 11.5 | 3.3 | 3.3 | 300 |
| 20 | Novo-4f | 13602496 | 7 | 18 | 42.7 | 3.6 | 4.0 | 875 |
| 21 | Novo-4 | 13602496 | 25 | 19 | 43.5 | 3.6 | 4.0 | 875 |
| 22 | Novo-4 | 13602496 | 30 | 20 | 37.2 | 3.6 | 4.0 | 875 |
| 23 | Novo-4 | 13602496 | 52 | 21 | 37.2 | 3.6 | 4.0 | 875 |
| 24 | Novo-4 | 13602496 | 66 | 22 | 39.2 | 3.6 | 4.0 | 875 |
| 25 | Novo-4 | 13602496 | 69 | 23 | 44 | 3.6 | 4.0 | 875 |
| 26 | Novo-4 | 13602496 | 91 | 24 | 39.7 | 3.6 | 4.0 | 875 |
| 27 | Novo-4 | 13602496 | 94 | 25 | 39.4 | 3.6 | 4.0 | 875 |
| 28 | Novo-4 | 13602496 | 98 | 26 | 37.6 | 3.6 | 4.0 | 875 |
| 29 | Novo-4 | 13602496 | 123 | 27 | 41.8 | 3.6 | 4.0 | 875 |
| 30 | Novo-4 | 213 | 25 | 10 | 38.7 | 3.6 | 3.3 | 1625 |
| 31 | Novo-4 | 213 | 25 | 11 | 38.2 | 3.6 | 3.3 | 625 |
| 32 | Novo-4 | 213 | 25 | 12 | 38.2 | 3.6 | 3.3 | 1125 |
| 33 | Novo-4 | 213 | 25 | 8 | 21 | 3.6 | 3.3 | 2285 |
| 34 | Novo-4 | 213 | 25 | 9 | 27.6 | 3.6 | 3.3 | 125 |
| 35 | Novo-4 | 213 | 63 | 16 | 32.6 | 3.6 | 3.3 | 1875 |
| 36 | Novo-4 | 213 | 63 | 17 | 33.1 | 3.6 | 3.3 | 375 |
| 37 | Novo-4 | 213 | 64 | 13 | 22 | 3.6 | 3.3 | 2225 |
| 38 | Novo-4 | 213 | 64 | 14 | 20.4 | 3.6 | 3.3 | 125 |
| 39 | Novo-4 | 213 | 64 | 15 | 34.9 | 3.6 | 3.3 | 1625 |
| 40 | Novo-4 | 13626135 | 65 | 182 | 22.86 | 3.592 | 12.4 | 100 |
| 41 | Novo-4 | 13626135 | 65 | 21 | 41.5 | 3.592 | 12.4 | 1000 |
| 42 | Novo-4 | 13626135 | 65 | 69 | 31.32 | 3.592 | 12.4 | 2150 |
| 43 | Novo-4 | 13626135 | 67 | 149 | 41.9 | 3.585 | 12.4 | 1000 |
| 44 | Novo-4 | 13626135 | 68 | 162 | 44.2 | 3.585 | 12.4 | 1000 |
| 45 | Novo-4 | 13626135 | 69 | 135 | 29.9 | 3.592 | 12.4 | 100 |
| 46 | Novo-4 | 13626135 | 69 | 57 | 36.2 | 3.592 | 12.4 | 2150 |
| 47 | Novo-4 | 13626135 | 69 | 79 | 46.3 | 3.592 | 12.4 | 1000 |

[a]Shaded rows indicate samples located at periphery assembly locations or adjacent to the central instrument tube

[b]GWd/tU = Gigawatt-days per ton of uranium

[c]The time from the fuel was discharged from the reactor to when the measurement was done

[d]Sample location from the bottom of the active fuel region

[e]Novovoronezh-3

[f]Novovoronezh-4

**Table 3:** Measured VVER-440 spent fuel samples [16] [17] [18].

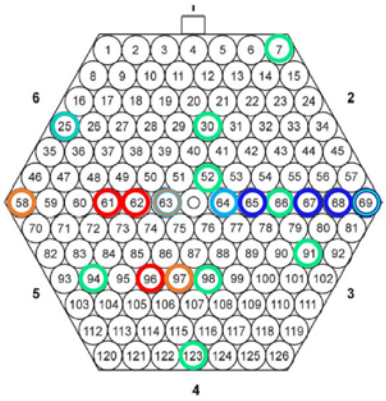

**Figure 2:** Schematic showing the arrangement of fuel rod identifiers in the VVER-440 assemblies and locations of measured rods; each color represents samples from a specific reactor. The assembly alignment/orientation notch is shown on side 1 (see the 1-6 numbers outside the assembly).

Problems were previously identified for the DA measurement of some nuclides from the Kola-3 assembly 144-46879 [19]. The present analysis of the data, previous independent analyses [19], and comparisons of the Kola-3 measurements with other similar VVER-440 fuels, suggest a systemic under-reporting of measured plutonium by ~ 18%, a value derived from the average bias in $^{238\text{-}242}$Pu for all 12 Kola-3 samples. This bias is observed in all isotopes of plutonium and is likely attributed to problems in the chemical separations process or the reference standards used in the measurements. Consequently, the data for all plutonium isotopes in these Kola-3 samples were adjusted to correct for the observed bias.

The comparisons of nuclide concentrations presented in this paper were focused to the nuclides of primary importance to spent fuel safeguards—namely, $^{235}$U, $^{238,239,240,241}$Pu, $^{244}$Cm, and $^{137}$Cs—although results for other nuclides, including $^{236}$U, $^{238}$U, $^{242}$Pu, $^{242}$Cm, and $^{134}$Cs, are summarized in Table 4.

The $^{235}$U results are shown in Figure 3. Note the same color coding as in Figure 2 is used here. The percentage values in parentheses in the legend are the initial enrichments of the samples. DA measurements are shown with error bars corresponding to the 95% confidence level, and calculations are shown as colored square symbols. Samples from the same assembly use the same color. The results show bands that are correlated to the different initial enrichments of the fuel rods—nominally 3.3, 3.6, and 4.4 wt % $^{235}$U. The calculated results reside within the measurement and uncertainty bands for most of the samples, and they trend consistently with burnup. There is a general trend to overestimate $^{235}$U in samples of the Novo-4 assemblies; this trend is not observed in Kola-3 or Novo-3 data. Many of the Novo-4 samples were taken from rods located at the periphery of the assembly, and larger deviations for these samples are expected. This overprediction is likely directly related to an overprediction in $^{239}$Pu for these same samples (see Figure 5) that is consistent with the rod position in the assembly. The periphery rods are generally subject to a more thermal, moderated neutron environment due to the additional water at the assembly gap, resulting in less plutonium production compared to the harder neutron spectrum in the assembly interior rods. These ORIGEN calculations were based on assembly average conditions, which are closer to the conditions experienced by the interior rods. Thus, the ORIGEN calculations will overpredict the plutonium concentrations for the Novo-4 samples from the periphery of the assembly. The overestimation of $^{239}$Pu leads to increased $^{239}$Pu fission, and consequently, it leads to less $^{235}$U consumption due to fission, resulting in an overestimation of the remaining $^{235}$U for these locations. For example, the Novo-4 sample with the highest burnup of 46.3 GWd/tU located at the corner of the assembly (rod #69) saw the largest overestimation of $^{235}$U, as represented by the rightmost blue square in Figure 3.

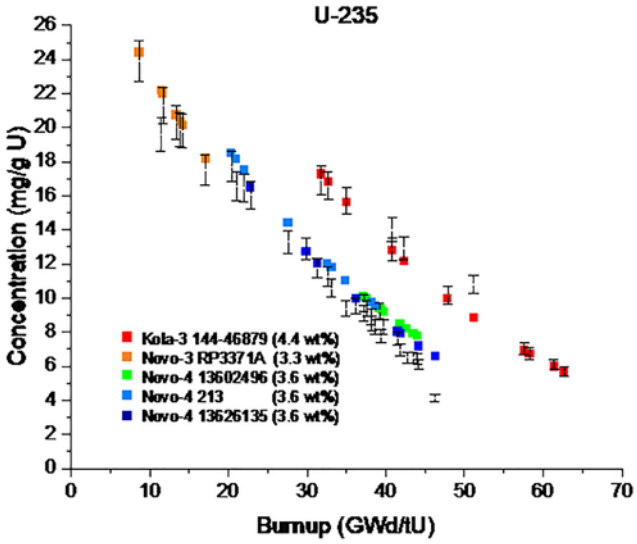


**Figure 3:** Comparison of measured (error bars) and calculated (color squares) $^{235}$U concentration in VVER-440 spent fuel samples.

The results for the plutonium isotopes $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, and $^{241}$Pu are shown in Figure 4 through 7, respectively. Note the same color coding as in Figure 2 is used in these figures. The calculated nuclide concentrations were in generally good agreement with measurements for all Pu isotopes. The overprediction of $^{239}$Pu in many of the Novo-4 samples was consistent with the fuel rod locations, as discussed previously. Note that the enrichment band positions for the $^{241}$Pu results are also affected by different cooling times of the measurements. As shown in Table 3, the Novo-4 13626135 samples had much longer cooling times than the other Novo-4 samples, which resulted in lower $^{241}$Pu concentrations in the Novo-4 13626135 samples than the other Novo-4 samples due to the extra decay of $^{241}$Pu, even though they had the same initial enrichments of 3.6%. Plutonium-241 has much shorter half life than the other three Pu isotopes studied here and it is more sensitive to differences in cooling times.

Results for $^{244}$Cm, the dominant neutron source in spent fuel with a relatively long cooling time (>2 years), are shown in Figure 8. Measurement uncertainties were generally much larger for curium isotopes than for the uranium and plutonium isotopes because of different measurement techniques used [20]. The results show that the calculations trend well with burnup and most of the measurement data. Accumulation of $^{244}$Cm in spent fuel is generally considered to trend to the ~4$^{th}$ power of burnup, which was consistent with the calculation results and measurement data included in this figure.

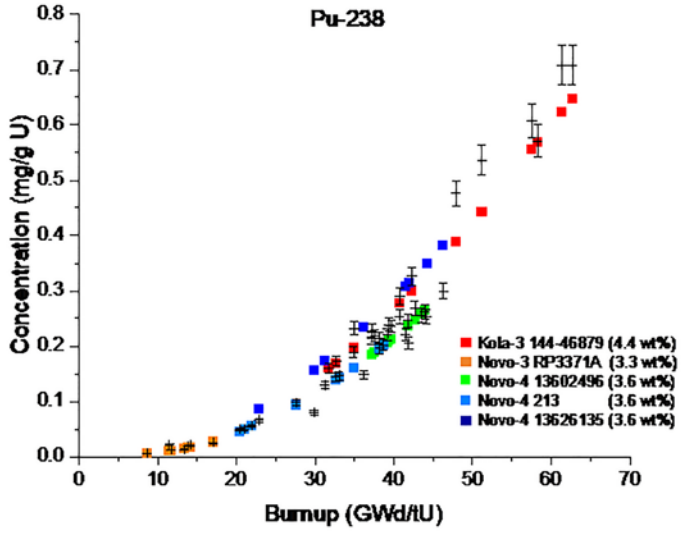


**Figure 4:** Comparison of measured (error bars) and calculated (color points) $^{238}$Pu concentration in VVER-440 spent fuel samples.

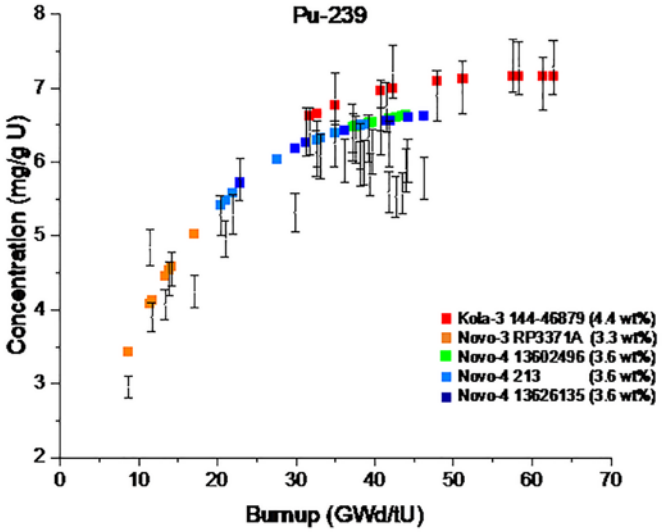


**Figure 5:** Comparison of measured (error bars) and calculated (color points) $^{239}$Pu concentration in VVER-440 spent fuel samples.

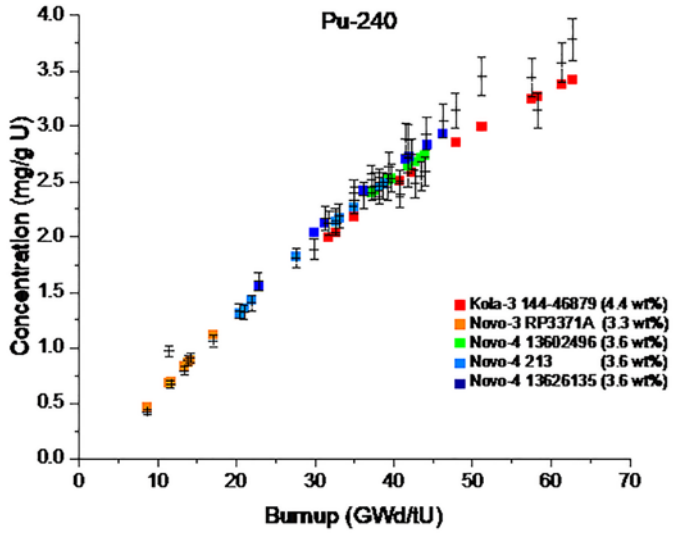


**Figure 6:** Comparison of measured (error bars) and calculated (color points) $^{240}$Pu concentration in VVER-440 spent fuel samples.

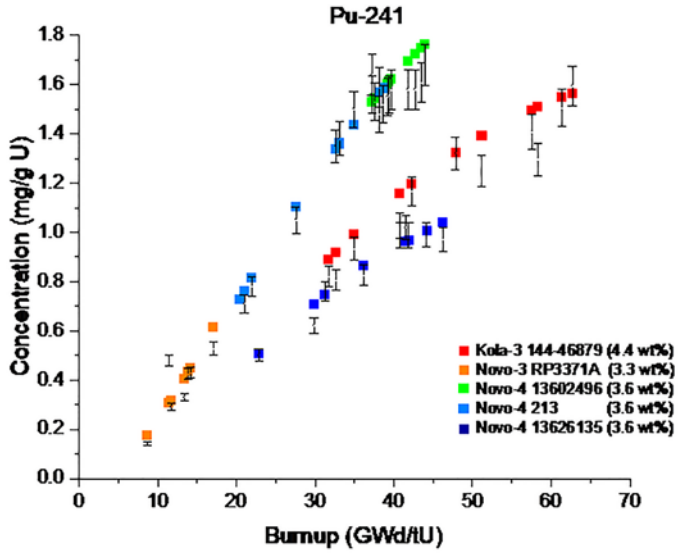


**Figure 7:** Comparison of measured (error bars) and calculated (color points) $^{241}$Pu concentration in VVER-440 spent fuel samples.

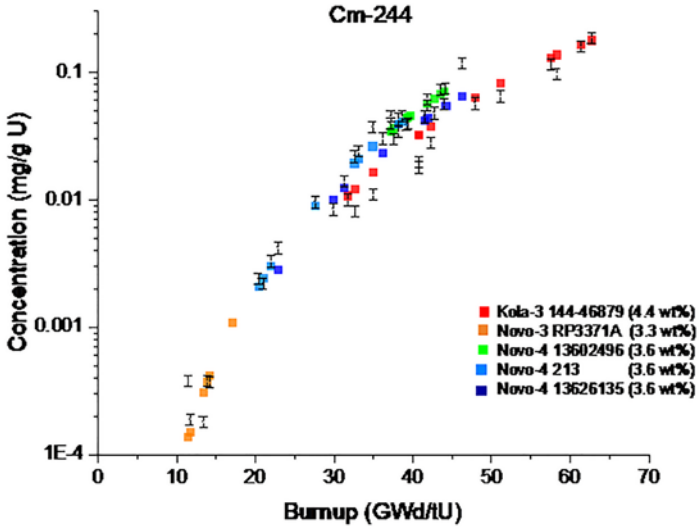


**Figure 8:** Comparison of measured (error bars) and calculated (color points) $^{244}$Cm concentration in VVER-440 spent fuel samples.

Fission products are the primary gamma emission sources in spent fuel, with the nuclides $^{137}$Cs, $^{134}$Cs, and $^{154}$Eu being dominant for gamma NDA measurements at longer cooling times (> 5 years). Measurements of $^{134}$Cs and $^{154}$Eu were available for the Kola-3 samples but were not considered due to likely errors in measured values from incorrect decay time corrections. Cesium-137 has a half life of ~30 years, which is much longer than that of $^{134}$Cs or $^{154}$Eu and is less sensitive to decay time corrections. The results for $^{137}$Cs using available data are shown in Figure 9. Calculated values were generally within the estimated relative measurement error of typically ±5%. As shown in Table 3, the Novo-4 13626135 samples had much longer cooling time than that of Kola-3, which resulted in less $^{137}$Cs in Novo-4 13626135 samples than in Kola-3 samples due to the extra decay of $^{137}$Cs.

To provide a quantitative assessment of ORIGEN library performance, the mean and standard deviation of the difference between measurements and calculations of nuclide concentrations are summarized in Table 4. Several samples were removed due to very large deviations compared to other samples in the measurement set. Specifically, Novo-3 sample RP-3371A-97-7 (#19) and Novo-4 sample 13626135-69-79 (#47) were removed from the statistical analysis since the deviations for many of the major actinides exceeded three standard deviations (3σ) for the population of data for the other samples.

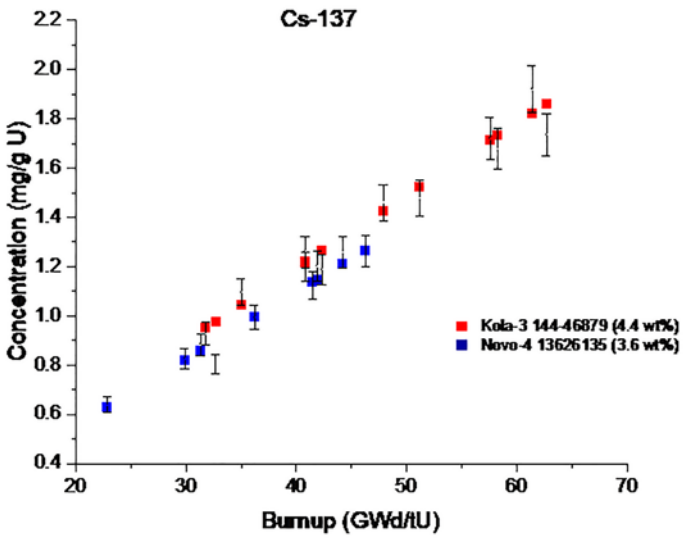


**Figure 9:** Comparison of measured (error bars) and calculated (color points) $^{137}$Cs concentration in VVER-440 spent fuel samples.

| Nuclide | Measurements # | Mean | Standard deviation |
|---|---|---|---|
| $^{235}$U | 45 | 6.1% | 8.9% |
| $^{236}$U | 45 | -3.1% | 6.0% |
| $^{238}$U | 45 | -0.4% | 0.3% |
| $^{238}$Pu | 38 | -5.9% | 8.2% |
| $^{239}$Pu | 44 | 5.7% | 6.3% |
| $^{240}$Pu | 45 | -0.8% | 5.1% |
| $^{241}$Pu | 44 | 5.3% | 7.0% |
| $^{242}$Pu | 44 | -0.9% | 8.9% |
| $^{242}$Cm | 12 | 1.2% | 29.5% |
| $^{244}$Cm | 43 | 7.5% | 27.3% |
| $^{134}$Cs | 19 | 14.4% | 20.7% |
| $^{137}$Cs | 19 | 0.8% | 6.2% |

Mean and standard deviation are calculated as and $\text{Mean} = \frac{1}{N}\sum_{k=1}^{N}\left(\frac{C}{M}-1\right)_k$ $\left\{\frac{1}{N-1}\sum_{k=1}^{N}\left[\left(\frac{C}{M}-1\right)_k-\text{Mean}\right]^2\right\}^{1/2}$, where N is total number of measurements, k is each sample, C is the calculated nuclide concentration, and M is the measured concentration.

**Table 4:** Statistical analysis of nuclide predictions.

The mean deviation between calculations and measurements (bias) was less than 8% for most nuclides, except for $^{134}$Cs probably due to its shorter half life and higher sensitivity to errors in decay time corrections. Similarly, the relative standard deviation (bias uncertainty) was less than 9% for the uranium and plutonium isotopes. Larger uncertainties were seen for the curium isotopes.

Calculations were performed using VVER-440 fuel assembly libraries that were developed to predict assembly average nuclide concentrations. However, measurements were performed on small samples from individual rods. Such inconsistency is avoided in a spent fuel safeguards practice because the NDA measurement and the ORIGEN calculations are performed on individual assemblies. Conversely, for predicting NDA signals using the ORIGEN modules, the uncertainties in the calculated nuclide concentrations contribute to only part of the overall uncertainties. Other factors, such as the detector response and multiplications in the assembly, also play a role, as discussed in the subsequent sections.

To assess the performance of the ORIGEN module's capability in predicting the FDET signals from VVER-440 assemblies, FDET measurement data were collected. FDET models were developed for these fuel types, and the calculated results of the ORIGEN module were compared to the measurement data, as discussed in the following three sections.

## 5. Loviisa measurement campaign

FDET measurements were performed on 13 VVER-440 assemblies at the Loviisa Nuclear Power Plant in February 2017 using a standard Euratom BWR FDET. The characteristics of the measured assemblies are summarized in Table 5. Assemblies from Loviisa units 1 and 2 were included in the measurements. All the measured assemblies operated in contiguous reactor cycles (i.e., they were not unloaded for one or more cycles before being reinserted in the core for further irradiation) except for assembly #1, which was out of the core during cycle 24.

## 6. FDET MCNP models for VVER-440

The FDET measurement configuration of the VVER-440 assemblies in the Loviisa spent fuel pool was modeled using MCNP to calculate the expected FDET signals (neutron counts and gamma current) as a function of emitted neutron/photon particle energy in the fuel. This calculation requires modeling the fuel assembly, the FDET instrument, and the pool water, including the soluble boron concentration used in most pools containing PWR fuel like VVER-440. The boron concentration was based on criticality safety considerations and is not necessarily constant, although levels are usually within a well-defined range. The Loviisa pools operate with a boron level of 13–15 g boric acid ($B(OH)_3$) per kilogram of fresh water [3]. Consistent with previous studies of the PNAR instrument [3], an average value of 14 g/kg was used in the current work, corresponding to a 2,450 ppm boron concentration.

The MCNP models for the VVER-440 assembly measurement configuration and details of the neutron/gamma detectors of the FDET are illustrated in Figure 10. The FDET used in the Loviisa measurement campaign was designed for BWR assembly types and has an internal width (opening) of 168 mm and a depth of 178 mm. The VVER-440 assembly outer dimensions are 145 mm (flat-to-flat) by 167 mm (diagonal). To avoid jamming the assembly in the FDET opening, the measurements were performed with the assembly rotated as shown in Figure 10.

| ID | Fuel type | Enrichment (wt %) | Reactor cycles | Burnup (MWd/tU) | Cooling time (days) |
|---|---|---|---|---|---|
| 1 | BNFL modified | 3.8 | 22, 23, 25 | 38,342 | 5,390 |
| 2 | BNFL modified | 3.8 | 22, 23, 24, 25 | 43,685 | 5,390 |
| 3 | TVEL standard | 3.6 | 15 | 5,716 | 8,120 |
| 4 | TVEL standard | 3.6 | 13, 14, 15 | 38,710 | 7,917 |
| 5 | TVEL standard | 4.37 | 33, 34, 35, 36 | 50,166 | 1,343 |
| 6 | TVEL fuel follower | 4.0 | 34, 35, 36 | 41,114 | 1,343 |
| 7 | BNFL standard | 3.7 | 32, 33, 34 | 40,068 | 2,071 |
| 8 | TVEL standard | 3.6 | 19, 20, 21 | 39,339 | 6,832 |
| 9 | TVEL standard | 3.6 | 24, 25, 26 | 41,749 | 5,012 |
| 10 | TVEL standard | 3.6 | 10, 11, 12 | 38,233 | 9,044 |
| 11 | TVEL standard | 3.6 | 13, 14, 15 | 39,635 | 7,917 |
| 12 | TVEL standard | 3.6 | 22, 23, 24 | 39,763 | 5,733 |
| 13 | TVEL standard | 4.37 | 31, 32, 33, 34 | 48,568 | 980 |

**Table 5:** Characteristics of measured VVER-440 assemblies.

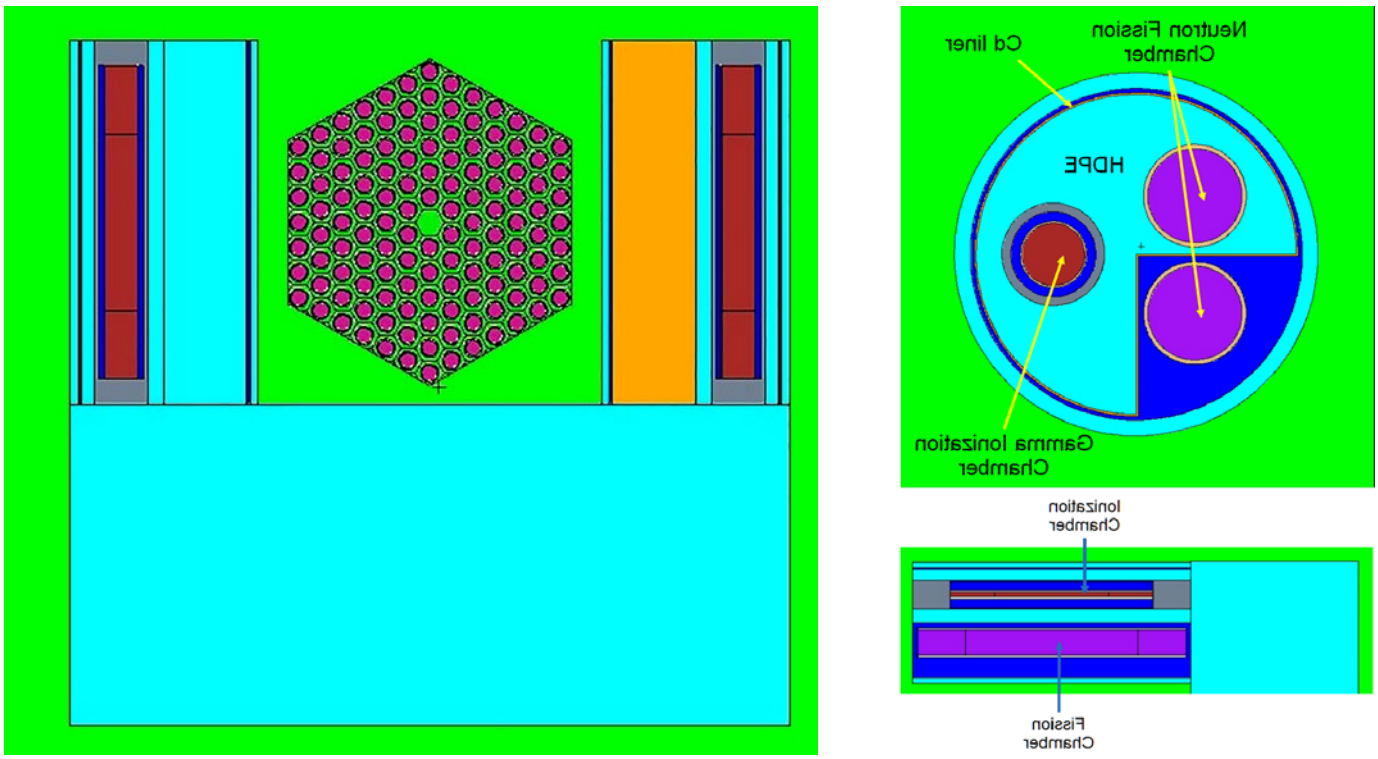


**Figure 10:** MCNP models of the FDET measurement of an VVER-440 assembly: (Left) the horizontal cross sectional view of the measurement configuration; (Right) the horizontal and vertical cross sectional views of one of the two tines of the FDET.

The FDET response functions were generated based on each of the neutron/photon source particle histories. For the FDET neutron response functions, 20 discrete source neutron energies ranging from 0.01–20 MeV were modeled, with one energy modeled in a separate MCNP model. Since most neutrons are born at ~2 MeV in spent fuel, the used neutron energy discretization was deemed sufficient. Similarly, the FDET gamma response functions and 18 discrete source photon energies ranging from 0.1–10 MeV were modeled. For the neutron/photon source particle energies that fell between the discretized energies, the response functions were interpolated by the ORIGEN module. In each MCNP model, a fixed-source calculation was performed using MCNP5 version 1.6 [7], with the source particles sampled uniformly in the horizontal direction of the fuel assembly but nonuniformly in the axial direction based on the calculated neutron/photon emission probability to account for the axial burnup variations. A number of 3E8 particle histories were used in the MCNP calculations, which took the neutron models 5,000–10,000 minutes to complete on a single processor, depending on which source energy was used, resulting in < 0.3% uncertainties in the calculated neutron response functions for most energies. For the photon models, it took 200–300 minutes to complete, resulting in < 1% uncertainties for the majority of energies. For the neutron signals, the fission rates in the $U_3O_8$ coating layers of the fission chambers were calculated, whereas the gamma dose rates in the active gas of the ionization chambers were calculated for the gamma signals. The calculated response functions are listed in Table 6.

| MeV | Gamma (×10$^{-10}$) |
|---|---|
| 0.1 | 0.001809 |
| 0.2 | 0.031539 |
| 0.3 | 0.092311 |
| 0.4 | 0.17800 |
| 0.5 | 0.27808 |
| 0.6 | 0.39391 |
| 0.7 | 0.51901 |
| 0.8 | 0.64721 |
| 0.9 | 0.78029 |
| 1.0 | 0.91562 |
| 1.2 | 1.18221 |
| 1.4 | 1.44747 |
| 1.6 | 1.68525 |
| 1.8 | 1.92923 |
| 2.0 | 2.15605 |
| 3.0 | 3.14516 |
| 5.0 | 4.68432 |
| 10.0 | 7.97987 |
| | |
| | |

| MeV | Neutron-A (×10$^{-3}$) | Neutron-B (×10$^{-3}$) |
|---|---|---|
| 0.01 | 0.73999 | 1.0554 |
| 0.03 | 0.76293 | 1.1244 |
| 0.06 | 0.77571 | 1.1668 |
| 0.1 | 0.78218 | 1.2083 |
| 0.2 | 0.79549 | 1.2756 |
| 0.4 | 0.79545 | 1.3309 |
| 0.6 | 0.79105 | 1.3850 |
| 0.8 | 0.77850 | 1.4052 |
| 1.0 | 0.77355 | 1.4042 |
| 2.0 | 0.72734 | 1.4108 |
| 3.0 | 0.67843 | 1.3437 |
| 4.0 | 0.64998 | 1.3084 |
| 5.0 | 0.59511 | 1.2095 |
| 6.0 | 0.59326 | 1.2092 |
| 7.0 | 0.59687 | 1.2046 |
| 8.0 | 0.61800 | 1.2332 |
| 9.0 | 0.60591 | 1.2156 |
| 10.0 | 0.60043 | 1.1999 |
| 14.0 | 0.61924 | 1.2397 |
| 20.0 | 0.67253 | 1.3303 |

Neutron-A = Fission chamber (bare)

Neutron-B = Fission chamber (with cadmium liner)

**Table 6:** FDET response functions for VVER-440 fuel.

## 7. Results of calculated and measured FDET signals

The FDET neutron/gamma signals for the 13 measured VVER-440 assemblies were also calculated using the ORIGEN module with the new VVER-440 libraries and the FDET response functions that were generated in this work. The module used the ORIGEN code to calculate the assembly average nuclide concentrations, the infinite neutron multiplication factor of the fuel ($k_\infty$) used to account for subcritical neutron multiplication, and the neutron/photon emission rates in each energy group from a given VVER-440 spent fuel assembly using the cycle-average irradiation histories provided by the operator. The module also combined the neutron/photon emission rates with the detector response functions to predict the FDET neutron/gamma signals for each fuel assembly. Before the predicted signals were compared against the measured ones, several additional data processing procedures were implemented, as discussed below.

Previous studies have indicated that the LND ionization chamber (Model 52110) used in the Euratom FDET exhibits a nonlinear response to the gamma dose rate [4]. To account for this, the calculated gamma signals in this work were further modified using an empirical correlation developed in the previous work [5] with a power coefficient of 0.77. The calculated neutron count rates were also modified to account for the neutron multiplication [5] of a given assembly using the infinite neutron multiplication factor for the fuel ($k_\infty$) calculated by ORIGEN and the neutron leakage factor ($L$) predetermined by MCNP for a particular fuel assembly design. The $L$ factor was set to 0.765 given the 2,450 ppm boron concentration in the pool; it would have been 0.626 if there were no boron in the pool.

An absolute calibration factor was not available for these assemblies. Instead, the ratio of the measured average signal to the calculated average was used to scale the calculated signals in each fuel assembly in the set to account for factors that were not considered (e.g., electronic efficiency, conversion of gamma instrument current to gamma signal) in the ORIGEN calculations or by the response functions. Calibration factors can be developed using measurement data collected by the same FDET instrument at the same spent fuel pool.

With these modifications, the calculated FDET neutron/gamma signals were compared to the measured values for all 13 assemblies. Table 7 summarizes these results and lists the calculated neutron multiplication of the assemblies as determined using the $k_\infty$ from ORIGEN and the $L$ factor from MCNP. The relative deviations between the measurements and calculated signals for each assembly are plotted in Figure 11. Note that the data acquisition software of the FDET used in this measurement converted the measured electric current from the two ionization chambers into digital signals using an arbitrary constant factor, which was accounted for using a detector calibration factor. The

calculated FDET signals were observed to be generally within ~ 10% of the measurements for all assemblies, except for assembly #3. The relative standard deviations for the deviations between calculations and measurements among all assemblies were 10.5, 10.8, and 5.6% for the neutron-A, neutron-B, and gamma signals, respectively.

Assembly #3 was only irradiated in the reactor for one cycle, and it achieved a very low discharge burnup of 5.7 GWd/tU. Due to the low burnup, the neutron count rates for this assembly (< 0.4 cps) are approximately three orders of magnitude lower than that of the other assemblies, so statistical counting uncertainties (~9%) may have contributed to the large error. (The counting time for these FDET measurements was ~5 minutes.) Further work to resolve the deviations for this assembly is ongoing. If assembly #3 were excluded from the set, then the relative standard deviations would be reduced to 5.8, 5.9, and 5.2% for the neutron-A, neutron-B, and gamma signals, respectively. These results are consistent with those reported previously for PWR measurement campaigns [5].

As with previous measurement campaigns, the correction for the gamma nonlinear ionization chamber response was found to be essential to the data analysis; assuming a linear response increases the relative standard deviation of the gamma results from 5.6% to more than 23%, with the largest deviations exceeding 45% for assembly #3

| | | | | | Cooling | Measurements (M) | | | Calculations (C) | | | | C/M-1 (%)[d] | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| FA # | Fuel type[a] | Profile type[b] | Enrich (wt %) | Burnup (MWd/t) | time (days) | nA (cps) | nB (cps) | gamma (arbitrary units[c]) | nA (cps) | nB (cps) | gamma (units) | Multiplication | nA | nB | gamma |
| 1 | 4 | F | 3.8 | 38,342 | 5,390 | 246.00 | 128.33 | 354,081 | 246.6 | 125.6 | 360,031 | 1.289 | 0.2 | -2.1 | 1.7 |
| 2 | 4 | F | 3.8 | 43,685 | 5,390 | 400.00 | 209.00 | 446,000 | 412.0 | 209.8 | 398,095 | 1.274 | 3.0 | 0.4 | -10.7 |
| 3 | 1 | F | 3.6 | 5,716 | 8,120 | 0.43 | 0.22 | 76,650 | 0.558 | 0.287 | 70,335 | 1.438 | 29.8 | 30.4 | -8.2 |
| 4 | 1 | F | 3.6 | 38,710 | 7,917 | 224.63 | 119.57 | 310,250 | 221.2 | 112.7 | 310,382 | 1.274 | -1.5 | -5.8 | 0.0 |
| 5 | 1 | P | 4.37 | 50,166 | 1,343 | 916.25 | 450.67 | 1,151,348 | 878.8 | 447.5 | 1,129,271 | 1.289 | -4.1 | -0.7 | -1.9 |
| 6 | 2 | F | 4.0 | 41,114 | 1,343 | 434.00 | 220.37 | 921,250 | 474.0 | 241.4 | 949,539 | 1.301 | 9.2 | 9.5 | 3.1 |
| 7 | 3 | F | 3.7 | 40,068 | 2,071 | 476.38 | 239.25 | 715,824 | 438.2 | 223.2 | 679,193 | 1.295 | -8.0 | -6.7 | -5.1 |
| 8 | 1 | F | 3.6 | 39,339 | 6,832 | 282.45 | 141.15 | 325,193 | 263.8 | 134.3 | 336,003 | 1.275 | -6.6 | -4.8 | 3.3 |
| 9 | 1 | F | 3.6 | 41,749 | 5,012 | 398.62 | 207.85 | 373,438 | 402.9 | 205.2 | 403,609 | 1.274 | 1.1 | -1.3 | 8.1 |
| 10 | 1 | F | 3.6 | 38,233 | 9,044 | 209.88 | 108.86 | 272,250 | 188.2 | 95.9 | 288,876 | 1.273 | -10.3 | -11.9 | 6.1 |
| 11 | 1 | F | 3.6 | 39,635 | 7,917 | 254.09 | 128.00 | 314,278 | 243.0 | 123.8 | 315,971 | 1.271 | -4.3 | -3.3 | 0.5 |
| 12 | 1 | F | 3.6 | 39,763 | 5,733 | 340.19 | 172.61 | 373,288 | 308.1 | 156.9 | 365,497 | 1.277 | -9.4 | -9.1 | -2.1 |
| 13 | 1 | P | 4.37 | 48,568 | 980 | 803.03 | 392.18 | 1,265,443 | 811.6 | 413.2 | 1,331,992 | 1.295 | 1.1 | 5.4 | 5.3 |

[a]1 = TVEL standard fuel; 2 = TVEL fuel follower; 3 = BNFL standard fuel; 4 = BNFL modified fuel

[b]P = profiled enrichment assembly; F = flat (uniform) enrichment assembly

[c]The FDET software converted the measured electric current from the gamma detectors into digital signals with a constant factor.

[d]nA = neutron-A (bare) channel; nB = neutron-B (Cd liner) channel

**Table 7:** Summary of VVER-440 Fork measurements and analysis results.

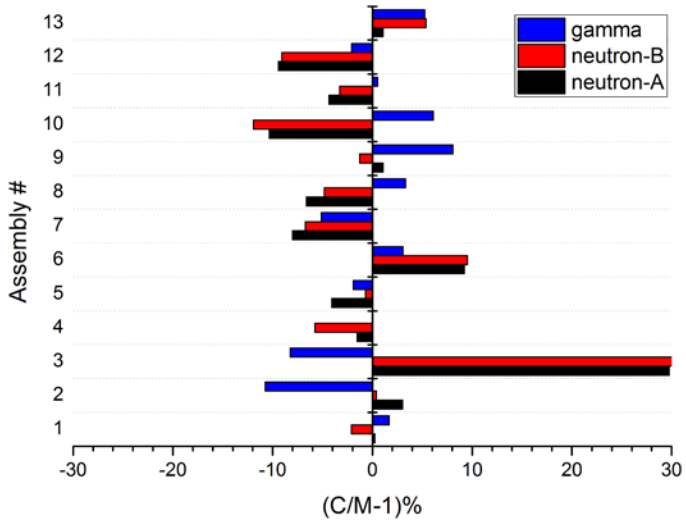


**Figure 11:** Relative difference between the calculated (C) and measured (M) neutron and gamma signals for the 13 measured VVER-440 assemblies.

(smallest signal) and assembly #13 (shortest cooling time and largest gamma signal).

Additional calculations were performed for assemblies #1, #2, #5, and #13 using ORIGEN libraries that were developed for both TVEL and BNFL/Westinghouse designs. Additional calculations were also performed to compare results using flat and different enrichment profile designs, since inspectors might not have access to this type of information. For these calculations, the effect of using different VVER-440 design libraries was < 1% for the gamma signal and 2–4% for the neutron signals. These differences are not large compared to other uncertainties and suggest that detailed knowledge of the fuel vendor or the detailed enrichment profile of the assembly are not essential.

Thresholds for significant discrepancies must be set by the safeguards inspectorate and the facility operator to trigger alarms when anomalies are suspected in the spent fuel assembly, the operator declarations, or the measurements. Careful considerations must be taken to set such thresholds to minimize the likelihood of false positive or false negative events. The thresholds must consider all the uncertainty sources in the predicted signals, including the uncertainties in the predicted nuclide concentrations and neutron/gamma source terms by the ORIGEN code with the ORIGEN libraries, the uncertainties in the response functions, the uncertainties in the measurements, the uncertainties caused by lack of detailed information regarding to fuel design and operating conditions, etc. These results provide initial data that can be used to develop such thresholds.

## 8. Summary and conclusions

The FDET (a mature technology) and the PNAR (a new technology) are expected to play a role in safeguards measurements at the Finnish spent fuel encapsulation plant for final disposal. The ORIGEN module, incorporated into IAEA and Euratom's IRAP software, was developed to predict the FDET signals using pre-generated ORIGEN libraries, response functions, and operator declarations to assist the safeguards inspectors in identifying anomalies in real time. The ORIGEN module can be modified to predict signals for other NDA (e.g., PNAR) spent fuel measurements.

This paper provides an analysis of FDET measurement data obtained during a VVER-440 measurement campaign at the Loviisa Nuclear Power Plant in Finland for 13 assemblies. The FDET signals of these 13 assemblies were calculated using the new ORIGEN libraries, the new response functions, and the operator declarations of the fuel assemblies provided by the Finnish reactor operator. The results show that the calculated FDET neutron and gamma detector signals are generally within 12% of the measurements except for one assembly. This assembly had a much lower burnup than the others, and it had a measured neutron count rate of only 0.4 cps. Therefore, the large discrepancy between the calculation and measurement in this case (~30%) can be attributed to poor counting statistics. If the assembly is excluded from the statistics, the average relative standard deviation between calculations and measurements is < 6% for both neutron and gamma signals. This result is consistent with previous results obtained for PWR measurement campaigns.

New ORIGEN libraries were developed using SCALE/TRITON based on ENDF/B-VII.1 nuclear data and fuel design information for the VVER-440 assemblies used in Finland. Performance of the new ORIGEN libraries was assessed by comparing the predicted nuclide concentrations using these libraries to DA-measured quantities obtained from literature. The comparisons of measured and calculated concentrations illustrate a generally good performance of the new ORIGEN libraries, with most calculated values being within or close to the assigned measurement uncertainties with several exceptions, which are discussed and explained in the paper. For application to spent fuel NDA, the simulations trend very closely to the measurements as a function of fuel burnup. New FDET response functions were generated using MCNP models for the VVER-440 fuel assemblies.

These results are directly applicable to preparing the ORIGEN module and FDET for use in safeguards verifications of spent VVER-440 fuel assemblies in routine inspections. These results are also applicable to future safeguards verifications in the planned Finnish encapsulation plants. A comprehensive study on various uncertainty components in the calculated FDET signals is recommended for future work to establish the acceptance threshold for the deviation between the calculated and measured FDET signals. The ORIGEN module is also being modified to predict PNAR signals in an ongoing effort.

## 9. References

[1] CHADWICK, M., HERMAN, M., OBLOŽINSKÝ, P., & al., e. (n.d.). *ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data.* Nuclear Data Sheets 112:12 (2011) pp. 2887-2996. https://doi.org/10.1016/j.nds.2011.11.002.

[2] Gauld, I., Giaquinto, J., Delashmitt, J., Hu, J., Ilas, G., Haverlock, T., & Romano, C. (2016). Re-evaluation of spent nuclear fuel assay data for the Three Mile Island unit 1 reactor and application to code validation. *Annals of Nuclear Energy, 87*, 267-281.

[3] Gauld, I., Hu, J., Baere, P. D., & al., e. (2015). In-Field Performance Testing of the Fork Detector for Quantitative Spent Fuel Verification. *37th ESARDA Annual Meeting, ISBN 978-92-79-49495-6.* Manchester, UK.

[4] Gauld, I., Hu, J., Baere, P. D., & al., e. (2015). In-Field Performance Testing of the Fork Detector for

Quantitative Spent Fuel Verification. *ESARDA, ISBN 978-92-79-49495-6.* Manchester, UK.

[5] Gauld, I., Radulescu, G., Ilas, G., Murphy, B., Williams, M., & Wiarda, D. (2011). Isotopic depletion and decay methods and analysis capabilities in SCALE. *Nuclear Technology, 174*(2), 169-195, https://doi.org/10.13182/NT11-3.

[6] GAULD, I., SLY, N., & MICHEL-SENDIS, F. (n.d.). OECD/NEA Benchmark Database of Spent Nuclear Fuel Isotopic Compositions for World Reactor Designs. *PHYSOR 2014.* Kyoto, Japan.

[7] HAVLUJ, F. (2011). *ISTC #3958 project benchmark specification and preliminary evaluation.* London, UK: presentation at the OECD NEA WPNCS meeting of the Expert Group on Assay Data for Spent Nuclear Fuel.

[8] Honkamaa, T., Levai, F., Turunen, A., Berndt, R., Vaccaro, S., & Schwalbach, P. (2014). A Prototype for Passive Gamma Emission Tomography. *Symposium on International Safeguards.* Vienna, Austria.

[9] Hu, J., Giaquinto, J., Gauld, I., Ilas, G., & Keever, T. (2017). Analysis of new measurements of Calvert Cliffs spent fuel samples using SCALE 6.2. *Annals of Nuclear Energy, 106*, 221-234.

[10] ILAS, G., MURPHY, B., & GAULD, I. (n.d.). VVER and RBMK Cross Section Libraries for ORIGEN-ARP. *Proc. of the 8th International Conference on Nuclear Criticality Safety (ICNC 2007), vol. 2, p. 413–417.* St. Petersburg, Russia.

[11] JARDINE, L. (2005). *Radiochemical Assays of Irradiated VVER-440 Fuel for Use in Spent Fuel Burnup Credit Activities.* Lawrence Livermore National Laboratory, UCRL-TR-212202. http://www.llnl.gov/tid/lof/documents/pdf/319467.pdf.

[12] KUOPANPORTTI, J. (2016). *Design and simulation of reactor operating cycle.* presentation for Aalto University, PHYS-E0562 Nuclear Engineering. https://mycourses.aalto.fi/pluginfile.php/456531/mod_folder/content/0/PHYS_E0562_Kuopanportti.pdf?forcedownload=1.

[13] MAKAROVA, T., BIBICHEV, B., & DOMKIN, V. (2008). Destructive Analysis of the Nuclide Composition of Spent Fuel of WWER-440, WWER-1000, and RBMK-1000 Reactors. *Radiochemistry, 50*(4), 414-426.

[14] MICHEL-SENDIS, F., GAULD, I., MARTINEZ, J., & al., e. (n.d.). SFCOMPO-2.0: An OECD/NEA Database of Spent Nuclear Fuel Isotopic Assays, Reactor Design Specifications, and Operating Data. *Annals of Nuclear Energy, 110*(https://doi.org/10.1016/j.anucene.2017.07.022), 779-788.

[15] Nuclear Energy Agency. (2011). *Spent Nuclear Fuel Assay Data for Isotopic Validation (State-of-the-art Report).* NEA/NSC/WPNCS/DOV(2011)5, Nuclear Energy Agency. https://www.oecd-nea.org/science/wpncs/ADSNF/SOAR_final.pdf.

[16] OECD/NEA Working Party on Nuclear Criticality Safety. (n.d.). *Expert Group on Assay Data for Spent Nuclear Fuel.* https://www.oecd-nea.org/science/wpncs/ADSNF/index.html.

[17] Okko, O., Moring, M., Honkamaa, T., Martinkka, E., & Hmalainen, M. (2018). GOSSER - Geological Safeguards and Security R&D Project in Finland - How STUK prepares itself for the Final Disposal in Finland. *ESARDA Bulletin, 56*, 2-5.

[18] PAVLOV, S. (2011). *Investigation of the VVER-440 Fuel Burnup and Nuclide Composition for the Use in Burnup Credit-Related Tasks (BUC-II).* Dimitrovgrad, Russia: Final Scientific Technical Report on Task 5, ISTC Project #3958p, Research Institute of Atomic Reactors (SSC RIAR).

[19] REARDEN, B., & JESSEE, M. (n.d.). *SCALE Code System.* ORNL/TM-2005/39, Version 6.2.2, Oak Ridge National Laboratory, Oak Ridge, Tennessee (2017).

[20] Smejkal, A., Regula, J., Nordquist, H., Longo, J., & Linnebach, R. (2014). A New Software Development Paradigm. *Symposium on International Safeguards.* Vienna, Austria.

[21] SUBLET, J.-C., KONING, A., FORREST, R., & KOPECKY, J. (n.d.). *The JEFF-3.0/A Neutron Activation File—EAF-2003 into ENDF-6 Format.* JEFDOC-982, Commissariat à l'Energie Atomique, France, (2003).

[22] TOBIN, S., PEURA, P., HONKAMAA, T., DENDOOVEN, P., MORING, M., & CHAMPAGNE, C. B. (2018). *Passive Neutron Albedo Reactivity in the Finnish Encapsulation Context.* STUK report (Nuclear Materials), ISBN 978-952-309-406-2.

[23] Vaccaro, S., Gauld, I., Hu, J., & al, e. (2018). Advancing the Fork detector for quantitative spent nuclear fuel verification. *Nuclear Inst. and Methods in Physics Research, A, 888*, 202-217.

[24] Vaccaro, S., Gauld, I., Hu, J., & al, e. (https://doi.org/10.1016/j.nima.2018.01.066, 2018). Advancing the Fork detector for quantitative spent nuclear fuel verification. *Nuclear Inst. and Methods in Physics Research, A, 888*, 202-217.

[25] X-5 Monte Carlo Team. (2003). *MCNP - A General Monte Carlo N-Particle Transport Code, Version 5.* Los Alamos, NM: Los Alamos National Laboratories (LA-UR-03-1987).