

NDA Verification of Spent Nuclear Fuel Prior to Geological Disposal with Passive Neutron Albedo Reactivity

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Abstract

Geological disposal of spent nuclear fuel is planned to begin in Finland in a few years. The disposal will start with spent BWR fuel from Olkiluoto 1 and 2 units operated by TVO. Prior to transporting of the fuel to the encapsulation plant and geological repository, all fuel assemblies will be verified in the wet storage facility where they are currently stored. The verification will be done in collaboration with the International Atomic Energy Agency (IAEA), the European Commission (EC) and Radiation and Nuclear Safety Authority of Finland (STUK) using one set of NDA verification instruments with their measurement results shared between all three inspectorates. The presently foreseen NDA methods will be Passive Gamma Emission Tomography (PGET), approved by the IAEA for safeguards inspections in 2017, and Passive Neutron Albedo Reactivity (PNAR). A PNAR instrument has been developed in a STUK-led collaboration for specifically the purpose of the verification of the BWR fuel at Olkiluoto. This paper summarizes the research and development on the PNAR instrument that has been conducted at STUK in preparation for the upcoming geological disposal. Since 2019, annual measurement campaigns have been held at the Olkiluoto spent fuel storage facility to assess the capabilities of the PNAR instrument. Over the course of these campaigns, more than 50 different fuel assemblies have been measured, several of them in multiple campaigns. These measurements have demonstrated the PNAR's ability to verify the fissile material content of a spent fuel assembly. In addition, the PNAR's reactivity measurement correlates with the leftover reactivity of the fuel, which can be estimated through simulations when the history of an assembly is known. In the 2022 measurement campaign, an ORIGEN (Oak Ridge Isotope Generation and Depletion) module for PNAR developed by the EC and the Oak Ridge National Laboratory was used to simulate the measurement signals of PNAR for the verified fuel assemblies. Such methods will allow for on-site verification of operator declarations and be part of the PNAR verification process that will be automated in the future.

Introduction

Geological disposal of spent nuclear fuel will start in Finland in a few years. Spent fuel will be transported from the interim storages to the encapsulation plant of Onkalo, operated by POSIVA. There, the fuel is encapsulated into copper canisters, which are then transported into an underground geological repository, where the fuel will be buried in the Finnish bedrock. It will be extremely difficult to re-verify fuel items after disposal. Thus, one of the key aspects of safeguarding the disposal process is the non-destructive assay (NDA) verification of all fuel assemblies prior to encapsulation and maintaining the continuity of knowledge after verification.

The disposal process begins with the BWR fuel from Olkiluoto 1 and 2 units operated by TVO. Verification will take place at the Olkiluoto wet interim storage facility. Currently, one measurement station is planned, and the measurement data will be shared between the International Atomic Energy Agency (IAEA), the European Commission (EC), and the Finnish Radiation and Nuclear Safety Authority (STUK).

The presently foreseen NDA methods for verification will be Passive Gamma Emission Tomography (PGET), approved by the IAEA for safeguards inspections in 2017, and Passive Neutron Albedo Reactivity (PNAR). These methods have been chosen to complement each other and offer a unique and hard-to-trick verification system.

This paper will summarize the research and development (R&D) work on PNAR that has led to the current realization of the instrument to be deployed. PNAR is a passive NDA method that uses the neutron radiation of spent nuclear fuel to assay the multiplication of the measured item. STUK's current version features four measurement pods arranged around the measured fuel assembly. Each pod incorporates total neutron and gamma detectors identical to those used in FORK detectors (FDET), and is independently connected to a counting electronics unit, likewise identical to those used in FDET. Additionally, and unlike in FDET, a separate cadmium liner is introduced to modify the neutron albedo in the system. The Cd liner can be moved into the narrow gap between the fuel assembly and the measurement pods at will, creating two different measurement conditions referred to as high multiplying and low multiplying setups. The ratio of the measured fast neutron signals between these two setups, called the PNAR Ratio, is a measure of the neutron multiplication of the measured fuel assembly. Neutron multiplication is an attribute of fissile or fissionable material, allowing the PNAR to directly assay the nuclear material content of the measured item. In addition to the PNAR Ratio, the total neutron and gamma signals can be used to verify the operator declaration of burnup and cooling time of the fuel assembly.

For a more detailed description of the PNAR method, please refer to [1, 2].

History of the PNAR method

The concept of passive neutron assay of spent nuclear fuel while changing the system's reactivity was first proposed in a 1982 report by Lee and Lindquist [3]. The currently realized measurement system follows the principle suggested already 40 years ago. Multiple publications have explored and improved the PNAR method through the years. These include publication by Menlove and Beddingfield in 1997 [4], who incorporated time-correlated neutron double and triple rates into the PNAR measurement, and by Conlin and Tobin in 2011 [5], who used MCNP simulations to develop PNAR response functions based on a simulated spent fuel library. In addition to verifying spent nuclear fuel, the PNAR method has also been suggested for other applications, such as measuring the fissile content

of electrochemical recycle materials by LaFleur et al. in 2014 [6] and fingerprinting fuel assemblies for shipper/receiver matching for safeguards purposes by Evans et al. in 2010 [7].

To the authors' knowledge, two PNAR prototypes for spent fuel measurements have been built before STUK's version. The first prototype was designed for concept testing for MOX fuel of the heavy water moderated Fugen reactor in Japan. Instead of a replaceable cadmium liner, the prototype had two sets of neutron detectors embedded in polyethylene, half of which were also covered by a cadmium layer to reduce neutron albedo. A set of measurements on seven assemblies were conducted in 2015. Although making the high and low multiplying measurements simultaneously effectively halves the required measurement time, multiple hours were still needed to achieve statistically significant results. Two main contributors to the long measurement times were a large water gap between the fuel and the instrument and inherently low multiplication of the fuel. [8]

The second prototype was tested in Clab, Sweden, in 2018. The PNAR measurement instrument was actually a retrofitted cadmium liner on a DDSI (Differential Die-Away Self-Interrogation [9]) equipment. Because the Cd liner was a separate piece of hardware from the other equipment, its relative positioning with respect to the detectors changed between measurements, and the fuel had to be moved away from the instrument between measurements to remove the liner. Furthermore, half of the detectors were unusable due to conduit failure. The results of the PNAR prototype measurements have not been published. [10]

In the ASTOR (Application of Safeguards to Geological Repositories) group of experts' final report in 2017, recommendation was given for criteria for NDA verification of the integrity of spent fuel items [11]. Furthermore, the NDA system should verify the consistency between the fuel item and its declaration. Based on the report, the NDA system should be:

- capable of pin level detection,
- capable of verifying that the declared assembly is consistent with measured signals,
- capable of measuring assembly neutron multiplication,
- capable of measuring fuel assemblies at the measurement location and in the medium of interest,
- robust, low maintenance and have a low false alarm rate,
- difficult to trick with pin substitution and
- able to measure the weight of the assembly.

STUK used the recommendations of the ASTOR final report as a design basis when they started to design a suitable NDA verification system for the Finnish concept for nuclear safeguards of geological repositories [12]. The national concept states that the best available technology should be used. The currently foreseen methods to be utilized for the verification prior to geological disposal include NDA verification with PGET and PNAR, accompanied with a weight measurement. Together, these methods fulfil both the national requirements, as well as all the criteria suggested by the ASTOR group.

While the PGET's ability to detect missing fuel pins had already been demonstrated in 2014 [13], a demonstration of PNAR's ability to measure assembly multiplication in a consistent way and in a reasonable measurement time had not been conducted by 2017. A STUK-led R&D collaboration with Encapsulation NDA services, the Helsinki Institute of Physics and EC was started to develop a PNAR design suitable for NDA verification of spent nuclear fuel in Finland prior to geological disposal. By the end of 2017, a conceptual design was ready [1, 14]. The design was optimized through thorough MCNP simulations for e.g., optimal neutron count rate, gamma shielding and moderator thickness.

STUK decided to build a prototype PNAR instrument based on the conceptual design. The prototype was especially designed for spent fuel from Olkiluoto 1 and 2 BWR units. The construction of the prototype started in 2018 and was finished the following year [2]. The first measurement campaign was performed in 2019 in Olkiluoto spent fuel storage. There, PNAR's ability to quantify the multiplication of spent fuel in a repeatable manner was demonstrated [15]. Since then, annual measurement campaigns have been conducted to quantify the limits of the PNAR instrument and to build expertise before the upcoming disposal process starts. The results from the 2019-2021 measurement campaigns have been reported in [15] and [16]. The PNAR instrument is shown in Figure 1.

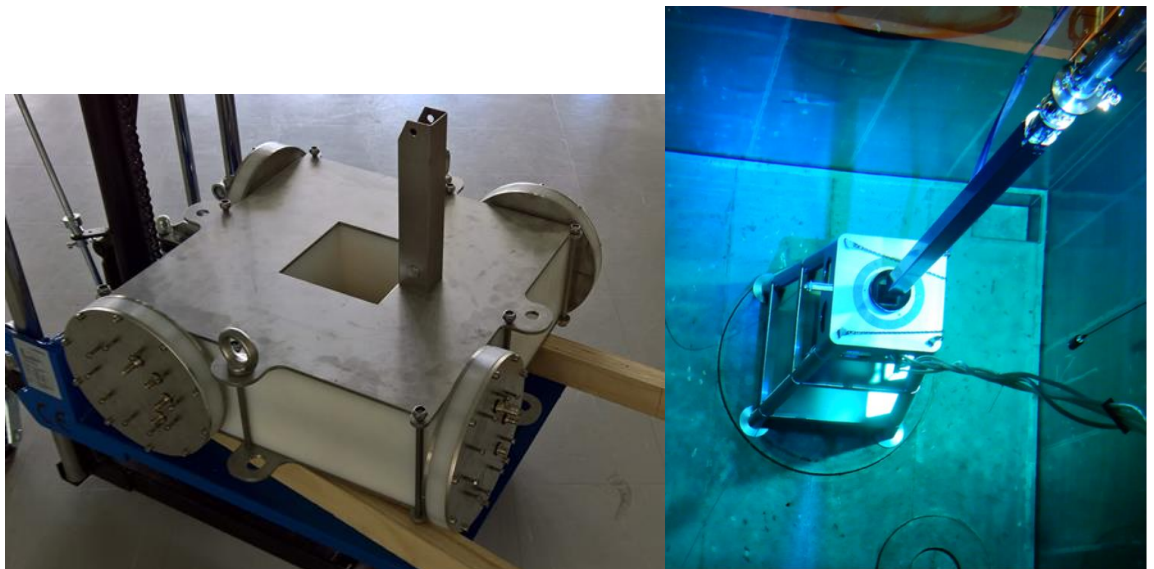


Figure 1: Left: PNAR prototype before mounting to its support structure. [Picture: STUK] Right: Fuel assembly being lowered into the measurement position in the Olkiluoto wet storage. PNAR is located below the top plate of the support structure seen in the picture. [Picture: TVO]

PNAR signal simulations of measured fuel assemblies

As suggested by the ASTOR final report, the NDA system for fuel verification prior to disposal should be able to verify the consistency between a fuel assembly declaration and the signals measured from the assembly by the system. One approach for accomplishing this objective is to use the declaration as input data and simulate the detector responses based on those. By comparing the simulated and measured signals, and given a pre-determined acceptance threshold, one can state whether the declaration and measurement agree within a given confidence interval.

The PNAR responses to given spent fuel assemblies can be simulated with the ORIGEN Data Analysis Module. Originally developed to simulate FORK signals [17], the ORIGEN Module is an ORIGEN (Oak Ridge Isotope Generation and Depletion) based burnup analysis code integration into the IRAP (Integrated Review and Analysis Package) software used by EC and the IAEA. The ORIGEN Module was expanded to include PNAR analysis under Action Sheet 65 between US DoE and EC. Under the AS-65, spent fuel assemblies originally measured in the 2019 Olkiluoto measurement campaign were later analysed using the ORIGEN Module and reported by Hu et al. in [18] and [19]. These publications also provide more detailed information on how the ORIGEN Module operates and how the simulated PNAR signals are generated.

In the most recent PNAR measurement campaign, held in August 2022, the ORIGEN Module was utilized already during the campaign. Simulated PNAR responses were generated before the measurements, based on a list of fuel assemblies chosen by STUK, and their accompanying declarations provided by TVO before the campaign. The trends between measured and simulated PNAR signals were then followed on site during the measurements. In addition to the traditional safeguards declaration (i.e., initial enrichment, burnup and cooling time), TVO also provided detailed reactor histories and their own SNF calculations about isotope fractions of the spent fuel assemblies.

The measurement campaign setting was similar to the foreseen verification case before disposal. There, POSIVA will provide TVO a list of assemblies to be disposed of and TVO will update their calculations of those assemblies. The updated declaration and the loading order of assemblies is sent to the inspectorates in advance. Thus, it is possible to have the simulation results available when an assembly arrives to the NDA measurement position. Then it is easy for an inspector on site or via remote access, or a completely automated system to accept or reject the verification results and send a corresponding signal to the operator.

Ten fuel assemblies were measured in the 2022 campaign. Their characteristics and the measured PNAR signals are listed in Table 1. For nine of those, simulated neutron multiplication along with gamma and neutron signals, with and without the Cd liner, were calculated using the ORIGEN Module. For each assembly, separate simulations were run with three different sets of input parameters. The input parameter sets were:

- Safeguards data, using only initial enrichment, burnup and cooling time,
- Operator data, with additional knowledge of local moderator density and burnup related to the axial measurement location of the assembly,
- Detailed operator data, with detailed power history of the assembly and knowledge of the axial moderator density. Assembly average burnup was used.

Safeguards data and operator data inputs correspond to the similarly named input data in [18]. For unknown parameters, typical values were used.

The net multiplications of all measured assemblies with different ORIGEN input parameter sets are compared against the measured PNAR Ratios in Figure 2. Multiplication trendlines are also shown for safeguards and operator input parameters from [18], where a larger set of assemblies was analysed. The datapoints with high multiplication at 1.039 PNAR Ratio correspond to assembly #1, which is an initial core assembly with significantly different operating history compared to an average assembly. This assembly should thus be treated as

a special case. Net multiplications calculated using the operator input data are lower than using the other two input data sets. Between the other two sets, safeguards data tends to result in slightly lower net multiplication than the detailed operator data. On average, the difference in net multiplication was 0.67 %. Detailed operator data includes knowledge of off-reactor cycles (i.e. the assembly has cooled for a longer period and then returned to the core). There was only one such assembly in the measured set (#28). This assembly had 1.1 % net multiplication difference between the safeguards and the detailed operator data sets.

The difference between the operator data and the other two input parameters is likely caused by the fact that the operator data set uses the burnup related to the axial position of the measurement, while the other two use assembly average burnup. Spent BWR assemblies typically have a high gradient in the axial burnup compared to PWR assemblies. For the measured assemblies, estimated burnup at the measurement height was on average 15 % higher than the assembly average burnup.

Table 1: Measured and/or simulated fuel assemblies in 2022 measurement campaign. The reported neutron and gamma signals are the measured ones without the Cd liner.

ID	IE (%)	Type	Burnup (MWd/tU)	Cooling (years)	Neutron (cps)	PNAR Ratio	Gamma (a.u.)
16	3.22	9x9-1AB	29200	28.3	-	-	-
70	3.23	9x9-1AB	35700	24.2	6400	1.043	250000
49	3.56	ATRIUM10	49700	14.2	27700	1.029	460000
71	3.56	ATRIUM10-9Q	40800	14.2	12500	1.046	380000
1	1.94	8x8-1	18600	38.3	600	1.039	90000
72	2.99	SVEA-64	36300	24.3	9000	1.042	280000
28	2.99	SVEA-64	32600	24.3	5700	1.046	230000
9	2.99	SVEA-64	37500	25.3	8000	1.039	260000
4	2.98	SVEA-64	37600	24.3	9800	1.046	290000
43	3.24	GE12	43100	15.2	16900	1.039	380000
46	3.51	GE14	43300	9.3	21800	1.048	520000

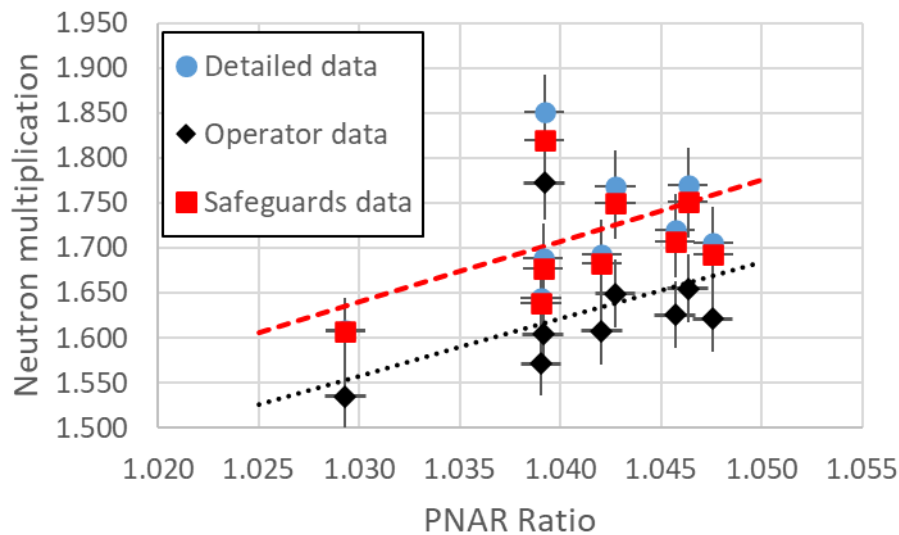


Figure 2: Simulated neutron net multiplication compared to measured PNAR Ratio. ORIGEN simulation results are shown using three different input data sets. The dashed lines are the safeguards (long dash) and operator (dotted) input data trendlines from [18], where a larger data set was analysed.

Conclusions and outlook

Although originally conceptualized in the 1980's, the first practical PNAR instrument for assaying the neutron multiplication in spent nuclear fuel has been deployed by STUK only in the recent years. The instrument was developed to accompany PGET in the final NDA verification of spent fuel prior to geological disposal that is beginning in Finland in a few years. The current PNAR realization allows for a short measurement time of approximately 5 minutes, has a robust design and is comprised of independent identical detector pods that will ease maintenance. The PNAR instrument has been tested in annual campaigns since 2019. In the most recent campaign in 2022, an ORIGEN Module for PNAR, developed by EC and ORNL, was utilized on site to predict the different signals measured by PNAR.

On its own, PNAR can already assay whether a fuel assembly contains fissile material and give a measure of its neutron and gamma emission rates and neutron multiplication. Paired with the ORIGEN Module, the system can be used to verify the correctness of the operator declaration of the assayed assembly. Although only a few assemblies were measured in the 2022 campaign, the relationship between the simulated neutron multiplication and the measured PNAR Ratio followed the same trend seen in earlier research. Further data, both from future measurements and simulations of earlier measurements, are needed to establish expected relationships between quantitative simulation and measurement results.

The disposal process in Finland is expected to last up to a hundred years and more advanced NDA methods will surely be established already in the first parts of that time frame. However, the robust PNAR method is already ready for deployment. The instrument was designed to fulfil the ASTOR recommendations together with PGET, and its detector and electronics designs are based on the well-established FORK detector, for which also the ORIGEN Module was originally developed. Utilizing the ORIGEN Module allows for easy interpretation of PNAR's verification measurement, which could also easily be automated if desired.

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