Utility of Including Passive Neutron Albedo Reactivity in an Integrated NDA System for Encapsulation Safeguards

S. J. Tobin¹, P. Peura², C. Bélanger-Champagne², M. Moring³, P. Dendooven², T. Honkamaa³

¹Encapsulation Nondestructive Assay Services, Los Alamos, NM 87544, USA. ²Helsinki Institute of Physics, FI-00014 University of Helsinki, Finland. ³Radiation and Nuclear Safety Authority - STUK, 00881 Helsinki, Finland. E-mail: steve.tobin.nm@gmail.com, p.peura@iaea.org

Abstract:

In August of 2017, the International Atomic Energy Agency (IAEA) issued the "Application of Safeguards to Geological Repositories (ASTOR) Report on Technologies Potentially Useful for Safeguarding Geological Repositories." In this IAEA report, the nuclear safeguards experts convened made recommendations on various aspects of encapsulation facility and repository safeguards. Specific to the non-destructive assay (NDA) requirements, the ASTOR experts made six specific recommendations. To satisfy these recommendations, a team working under the direction of the Finnish Radiation and Safety Authority researched the capability of an integrated NDA system that combines the capabilities of a Passive Gamma Emission Tomography (PGET) instrument, a Passive Neutron Albedo Reactivity (PNAR) instrument and a load cell. The current study focuses a conceptual design of the PNAR instrument capable of supporting several of the IAEA recommendations. To enable this research goal, the performance of a PNAR instrument, designed to measure boiling water reactor assemblies, was simulated using fuel with the isotopic content representative of fuel of various initial enrichments, burnups and cooling times. The research results illustrate the capability of the PNAR instrument to fulfil the IAEA recommendations while using robust, relatively simple, hardware.

Keywords: non-destructive assay, encapsulation safequards, PNAR

Introduction 1.

In August of 2017 the International Atomic Energy Agency (IAEA) issued the "Application of Safeguards to Geological Repositories (ASTOR) Report on Technologies Potentially Useful for Safeguarding Geological Repositories" [1]. For the formulation of this report, the IAEA convened groups of experts on specific topics to provide recommendation on various aspects of encapsulation facility and repository safeguards; this current study focuses on the Finnish implementation of the ASTOR Group recommendations in the context of a non-destructive assay (NDA) system. The mandate of the ASTOR NDA Focus Group was to improve upon the state-of-the-practice given the extremely difficult to access nature of fuel placed in a deep geological

repository. The NDA system proposed by Finland satisfies all the recommendations set forth by the NDA Focus Group by integrating a Passive Gamma Emission Tomography [1, 2, 3, 4] instrument with a Passive Neutron Albedo Reactivity (PNAR) instrument [1, 5, 6, 7]. The purpose of the current study is to describe how the PNAR instrument helps strengthen the NDA system in the implementation of the IAEA recommendations while using robust, relatively simple, hardware that can measure the assembly multiplication.

Requirements for the encapsulation NDA 2. system

Below is a list of suggested characteristics for the NDA system of a spent fuel assembly encapsulation facility. The list was created by the NDA Focus Group convened by the IAEA as part of ASTOR.

- **a.** Capability to detect individual pins, even though it is recognized that pin level detection might not be possible for all assembly fuel types and for all burnup and cooling time scenarios.
- b. Capability to verify that the declared assembly is consistent with measured signatures: Enough information is provided in the declaration of each assembly to predict, within useful limits, some measurable signatures from each assembly. Once predicted, a comparison between expectation and measurement is possible and recommended.
- c. Capability to measure assembly neutron multiplication: The neutron multiplication of an assembly can be measured with the neutron signal. Furthermore, this multiplication can also be calculated from the declaration. Multiplication is singled out in this list for its close connection to the presence of fissile material and because it is a bulk property of the assembly.
- d. Robustness, low maintenance and low false alarm rate must all be properties of the NDA system. The NDA system should not significantly impede facility operation; duplicate systems are recommended in addition to using robust technology.
- e. System should be difficult to trick with pin substitution. As noted in Chapter 1 of [1], "all individual NDA techniques ... can be tricked by a well-designed pin replacement." Hence, the aggregate NDA system needs

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to make well-designed pin replacement extremely difficult to plausibly/usefully perpetrate.

f. Capability to measure the total weight: A measurement of the assembly weight is considered relatively simple and able to contribute one more constraint a 'would be' proliferator needs to satisfy in designing a diversion scenario.

Fulfilling all the characteristics listed above will require an integrated NDA system. The focus of this paper is to describe how the PNAR instrument contributes to fulfilling characteristics (b) through (e).

For characteristic (a) Passive Gamma Emission Tomography (PGET) instrument is expected to provide pin level detection capability in Finland. Recent research on the NDA system intended for the Finnish encapsulation facilities indicates that detection of every pin in a boiling water reactor (BWR) assembly should be straightforward, while detection of every pin in a VVER-440 assembly, is a topic of ongoing research in Finland and at the IAEA [2, 3].

3. Passive neutron albedo reactivity physics

The PNAR NDA technique involves the comparison of the neutron count rate for an object when that object is measured in two different setups. One setup is designed to enhance neutron multiplication while the other setup is designed to suppress it. As implemented for the Finnish BWR fuel, the high multiplying section was produced by the assembly in the water of the pool, while the low multiplying section was created by putting 1 mm of Cd as close as possible to the fuel while it remained in the pool. Cd was selected due to its extremely large absorption cross-section for all neutron energies below ~0.5 eV. The PNAR signature is calculated by dividing the count rate measured in the high multiplying section by the count rate measured in the low multiplying section.

The first PNAR experiments with an assembly geometry were performed with a 15x15 fresh assembly; a ²⁵²Cf source was imbedded in the assembly to increase the neutron flux. The results showed a healthy change in the PNAR signature with changes in the average initial enrichment of the assembly [5]. An experiment using fresh rods in air was performed inside of a multiplicity counter showing that the sensitivity of a PNAR instrument will increase if the detectors efficiency is elevated enough to support correlated neutron detection [8]. The first use of a PNAR with spent fuel was performed with Fugen fuel. The main conclusion was that PNAR was able to discern levels of neutron multiplication; the results of the Fugen experiments were much less dynamic than is expected for typical commercial fuel setups because (1) the Fugen fuel contained little fissile material as the fuel was irradiated in heavy water, (2) the water gap around the fuel was approximately triple that which is expected for commercial fuel [9].

The PNAR implementation planned for Finland, an implementation that combines (a) a ³He detector tube and polyethylene surrounded by Cd and (b) a low multiplying section produced with a Cd-liner, lends itself to a conceptual discussion of the PNAR physics. The only significant difference in the measured count rate for a section of fuel measured in both the high and low multiplying sections, is the counts resulting from the multiplication caused by the neutrons that were absorbed in the Cd-liner. The counts produced by the neutrons not absorbed in the Cd-liner are in both the numerator and denominator of the PNAR Ratio so these high-energy neutrons that are unaffected by the Cd-liner create a PNAR Ratio of 1.0; any deviation from 1.0 is due to counts produced by chain reactions initiated by neutrons that are absorbed by the Cd-liner. Because the PNAR signal is produced by the neutrons returning into the fuel with an energy below the Cd-cutoff energy of ~0.5 eV, the PNAR technique is sometimes described as interrogating the fuel with low energy neutrons from the location of the Cd-liner.

There are two options for implementing the Finnish conceptual PNAR design: (1) either the Cd-liner is moved in and out of position to create the high and low multiplying setups, or (2) the fuel is moved between two detectors sections for which one section is high multiplying and the second section is low multiplying. The two sections in this later case could be identical in all ways except for the presence of a 0.5 mm thick sheet of Cd approximately 0.5 m long in the axial direction for the low multiplying section. This latter option can be implemented by putting the low and high multiplying sections on top of each other; in this case, the fuel is moved vertically about a meter between the two measurements to assure the same section of fuel is measured in both detector sections. The latter case has more flexibility to change the multiplication in that more than just Cd can be changed to differentiate between the two sections. In the case of Finland, moving the Cd-liner is expected. This has the benefit of requiring only one detector bank and the PNAR measurement can be completed without moving the fuel. As a result, the PNAR measurement can be completed during the same time as the PGET measurement.

4. Passive neutron albedo reactivity hardware

The Radiation and Nuclear Safety Authority (STUK) of Finland commissioned the conceptual design of a PNAR instrument as part of an NDA System designed to meet the safeguards and safety needs of Finland in the context of spent fuel encapsulation and geological disposal. The geometry of the PNAR instrument needs to be adapted to the dimensions of each fuel type. In the case of Finland, because different fuel types reside at different facilities, BWR-specific and VVER-specific designs were developed. In Figure 1 and Figure 2, two cross-cutting images of the conceptual design for BWR assemblies are illustrated. The size of some of the key components of the PNAR detector are the following: The ³He tubes are 17.4 mm in diameter with a fill pressure of 6 atm and an active length of 0.2 m. The lead, needed to reduce the gamma dose to the ³He tubes, is 52 mm thick at is thickest. All Cd layers are 1 mm thick. The Cd-liner surrounds the fuel and is 0.74 m long; shorter length liners are under investigation. The starting point for the design involved calculating the amount of lead needed to keep the gamma dose to the ³He tube below 0.2 Gy/hr limit for a 0.35 m active length tube [10]. The second step involved optimizing the polyethylene near the tube for the largest count rate possible. The final step involved making sure the count rate did not exceed the recommended maximum count rate of 5 x 10⁴ count/s for the selected tube [11].

In this section, the hardware used to implement the PNAR concept is described with an emphasis placed on how the PNAR design partially fulfils characteristic (d) of the ASTOR Experts Group, which recommends the use of robust, low maintenance hardware. The fulfilment of this recommendation is addressed while describing the key component of the PNAR instrument by comparing, when applicable, the hardware used in implementing the PNAR concept to that of a Fork detector [12, 13]. The Fork detector was selected because it is a robust safeguards instrument, which has been used in the field for several decades:

- With the inclusion of an ion chamber, the PNAR instrument as designed is effectively a high efficiency Fork detector with reduced positioning uncertainty due to (a) detectors located on each of the 4 sides of the fuel and (b) a smaller water gap between the fuel and the detector than with a Fork. Note that the second set of detectors, which are located above and below the assembly in Figure 2, are not visible because they are located 0.1 m below the illustrated detectors. STUK is currently investigating a PNAR design with all detectors on one axial level.
- 2. The key to implementing the PNAR concept is the "Cdliner" that is depicted between the fuel and lead section of the detector. Note that this Cd-liner is only present for one of the two PNAR measurements.
- A second layer of Cd, around the polyethylene, surrounds each ³He tube. This Cd ensures that the detector only detects epithermal and fast neutrons from the fuel; in some Fork detector designs, Cd is also used for this purpose. A more uniform spatial sensitivity across the assembly is achieved by detecting these higher energy neutrons [5].



Figure 1. Vertical cross-sectional view of the BWR PNAR detector along one side of a BWR fuel assembly. The lead shielding is 0.128 m vertically and 0.117 m horizontally. Proportions are accurate.



Figure 2. Horizontal cross-sectional view of the BWR PNAR detector relative to a 10x10 BWR fuel assembly, which is 0.126 m on a side. The detector units illustrated here span 0.128 m in the vertical direction below the vertical midplane of the detector. An identical set of detectors span the 0.128 m in the vertical direction above the midplane; these additional detectors look identical to those depicted here except they are rotated 90 degrees around the center of the fuel. Proportions are accurate.

There are three aspects of the PNAR hardware, as simulated, that deviate from the hardware of a Fork detector:

- ³He tubes are used instead of fission chambers. This selection was made to obtain the desired precision for typical fuel assemblies in two minutes or less. Fission chambers or boron tubes could be used if longer count times are acceptable. The lead is present to reduce the gamma dose to the ³He tubes; the lead would not be necessary if fission chambers are used; yet given that the installation is permanent, the weight of lead is not a significant concern.
- 2. The instrument is designed with the expectation that the fuel will be inserted from above into the detector. The detectors are located on the 4 sides of the assembly; it is expected that the instrument will not measure locations near the ends of the assembly. This selection was made to reduce the sensitivity to anisotropy in the assembly burnup. If such an uncertainty is not too large, a reduction to two detectors on opposite sides of the fuel is acceptable.
- 3. The presence of a 1 mm thick, 0.74 m long axial Cdliner. This sheet of metal is the sole PNAR component that is not necessary for a Fork detector.

5. Simulated passive neutron albedo reactivity signature

To access the capability of the PNAR detector to measure spent fuel, the PNAR ratio was calculated using the average isotopic content of 12 assemblies that span a range of initial enrichment (3, 4 and 5 wt.%), burnup (15, 30, 40 and 60 GWd/tU) and cooling time (20, 40 and 80 years) values. The Monte Carlo N-Particle Code (MCNP6[™]), Version 6 [14] was used for the PNAR simulations while the isotopic mixture of the various assemblies was produced by the Monteburns code [15] as part of the Next Generation Safeguards Initiative [16, 17].

Figure 3 shows the calculated PNAR ratio versus burnup for 12 different assemblies in fresh water. Two simulations were run to calculate each data point, once with the Cdliner in place and once without the Cd-liner. All data points in Fig. 3 are for fuel with a 20-years cooling time.

The PNAR Ratio values for all the data points with ratios above 1.1 were simulated in the standard manner, meaning that all neutrons, and subsequent reactions that they may cause, were followed until the neutrons were either absorbed or left the extremities of the simulation; any nuclear reactions that produced additional neutrons, such as induced fission, were followed through to fruition.

For the three assemblies considered to be nearly fully irradiated, given their initial enrichment and burnup values, which are the assemblies with PNAR Ratios of about 1.14, additional simulations were performed to calculate the PNAR ratio for the case when no induced fission could take place. The three assemblies are the following: (a) 3 wt.%, 30 GWd/tU, (b) 4 wt.%, 45 GWd/tU, (a) 5 wt.%, 60 GWd/tU; these assemblies are labelled separately in Fig. 4. For these 3 assemblies, induced fission reactions became absorption reactions. This is a useful exercise because it indicates the signal expected if all the fuel were replaced with a non-multiplying material. This change in the simulation was accomplished by adding the "NONU" card to the simulation. The calculated PNAR Ratio for each of these assemblies with the NONU card is 1.002, 1.003 and 1.008, respectively. The absolute value of the uncertainty on the PNAR ratio, propagated from the MCNP6[™] statistical uncertainty, is 0.003 for all points. The vertical extent of each data point in Figure 3 is approximately 4 times the propagated statistical uncertainty calculated with MCNP6[™].



Figure 3. the BWR PNAR Ratio, simulated with fresh water, is illustrated as a function of burnup for 12 assemblies of various initial enrichments and burnup values; 3 assemblies were simulated using the nonu card. The cooling time is 20 years. The vertical extent of each data point is approximately equal to 4-sigma of statistical uncertainty arising from the simulated statistics.

The following are key points concluded from Figure 3:

- 1. The change in the PNAR Ratio with increasing burnup is a smooth decreasing function of burnup for a given initial enrichment. If an assembly starts with more potential nuclear energy, it will be measured to have an elevated PNAR ratio when fresh.
- 2. Fully irradiated assemblies, regardless of their initial enrichment, are expected to have nearly the same PNAR Ratio.
- 3. The change in the PNAR Ratio between a fresh 4 wt.% assembly and a 4 wt.% assembly that was irradiated to 45 GWd/tU is approximately the same as the change in the PNAR Ratio calculated between a fully irradiated assembly and an assembly for which all the pins were replaced with non-multiplying material.

In Figure 4, the PNAR Ratio is graphed as a function of the "net multiplication;" which is calculated by the MCNP6 code by taking the ratio of the number of neutrons started in the fuel to the number of neutrons followed during the course of the simulation. Note that the net multiplication is calculated for the case of neutrons starting from all the pins in the assembly with the energy sampled from a Watt fission spectrum. The data points in Figure 4 include all the same data points illustrated in Figure 3 as well as 6 additional assemblies. These additional assemblies are for the three nearly fully irradiated assemblies already discussed; however, the isotopic content was "aged" to represent that which is expected for cooling times of 40 and 80-years, in addition to the 20-year cooling time case from Figure 3. The main point for including these assemblies is to show that the 9 fully irradiated assemblies occupy a small area of the overall parameter space; additionally, among these 9 assemblies, the 3 assemblies with the same initial enrichment but cooling times of 20, 40 and 80 years are clustered in an even smaller area.



PNAR Ratio for 10x10 BWR Assemblies vs Net Multiplication

Figure 4. the PNAR Ratio is graphed as a function of the net multiplication. 3 assemblies with cooling times of 40 years and 3 assemblies with cooling times of 80 years were included compared to Figure 3. All these longer-cooled assemblies group around a net multiplication value of 1.41 and a PNAR Ratio of 1.14 and are labeled as "fully irradiated assemblies."

The conclusions drawn from Figure 4 are the following:

- 1. Regardless of initial enrichment, burnup or cooling time, there is a smooth relationship between the PNAR Ratio and net multiplication.
- 2. There is a change of between 0.13 and 0.14 in the PNAR Ratio between any irradiated assembly and a non-multiplying assembly.
- 3. Almost all assemblies to be measured at an encapsulation facility will be fully irradiated. Hence, a near constant PNAR Ratio will be measured for all these assemblies; if the current simulations are representative, that value will be around 1.14.

4. From Figure 4, we can see that the impact of cooling time is relatively small, the 9 fully irradiated assemblies all have net multiplications values of around 1.4 and PNAR Ratios of around 1.14.

Because the uncertainty of the PNAR instrument is connected to how useful the instrument can be, the results from a separate report examining the anticipated uncertainty are summarized here [18]. In that report, the uncertainties due to the following were examined: (a) assembly position in the instrument, (b) counting statistics given a total measurement time of 5-minutes, (c) estimated uncertainty given non-uniform irradiation. The end conclusion was that a rough estimate of the 1-sigma uncertainty in the PNAR Ratio is anticipated to be +/- 0.005 for a typical BWR assembly (32 GWd/ tU, 40-year cooled) given a 5 minute total count time; while the coolest assemblies (17 GWd/tU, 60-year cooled) will likely need a 20-minute count time to obtain a similar uncertainty. To put the 0.005 value in some context, a 0.005 variation in the PNAR Ratio corresponds to the change in the multiplication caused by a burnup variation of 1.4 GWd/tU. Additionally, if a fully irradiated assembly were replaced with a non-multiplying assembly, then the PNAR Ratio should change from 1.142 to 1.002 for a net change of 0.140; this represents a change of 28 sigma; the main point being that a significant removal of fissile material from the assembly will be easily detected.

A point worth emphasizing is that the PNAR technique fulfils the recommendations of the IAEA ASTOR group that the NDA system be "capable of measuring assembly neutron multiplication."

6. Merit of an integrated NDA system

The ASTOR Experts Group recommendations (b) and (e) are discussed together within the context of the merits of an integrated NDA system. Characteristic (b) involves verifying the declaration with the measured signatures, while characteristic (e) involves creating an NDA system that is difficult to trick.

The integrated Finnish NDA system suggested by STUK has the following measured signatures:

- Relative distribution of the gamma ray emission within a horizontal cross-section of an assembly with pin level resolution using the PGET instrument. The PGET instrument [2], after recent refurbishment by IAEA [3], has shown a capability for automated detection of single or multiple missing pins in BWR and VVER-440 fuel. However, improved image reconstruction and analysis techniques are still required [4].
- 2. Absolute gross gamma intensity as measured by ion chambers, which are built into the PNAR instrument, and the absolute ¹³⁷Cs count rate as measured by the CZT detectors of the PGET instrument.

- 3. Absolute neutron count rate as measured with the PNAR detector and the boron-tube neutron detectors in the PGET instrument.
- 4. Absolute neutron multiplication as measured with the PNAR detector.

The analytic approach of the Finnish NDA system has two separate parts: (a) One part analyses the PGET information to create relative-intensity gamma-ray images. (b) The second analytic approach uses the information declared by the state to calculate the multiplication, absolute gamma and neutron source terms. Considerable research along these lines was performed by Euratom and Oak Ridge National Laboratory researchers [19]. Their analytic approach uses both SCALE [20] and MCNP6[™] to calculate the assembly multiplication as well as the neutron and gamma flux from the fuel to any relevant detectors.

Focusing on the SCALE/MCNP6[™] portion of the analysis for the Finnish case, the gamma intensity measured by both the CZT and ion chambers can be compared to the values calculated by SCALE and MCNP6[™] from the declaration. Similarly, the total neutron count rate and the neutron multiplication can be calculated from the declaration and compared to the measured values. In the case of the measured multiplication, either the PNAR Ratio itself could be calculated or a calibrated correlation between the net multiplication and the PNAR Ratio, as illustrated in Figure 4, could be used.

An additional possible part of the analysis could involve the calculation of the neutron source term. The neutron source term is equal to the total neutron emission divided by the net multiplication. Given the relationship between the net multiplication and the PNAR Ratio illustrated in Figure 4, the intensity of the neutron source term can be calculated.

In summary, ASTOR characteristic (b), which involves verifying the State declaration, will be satisfied for the longcooled fuel of interest in Finland by measuring the following characteristics of the fuel:

- 1. Both the absolute gross gamma intensity and ¹³⁷Cs count rate will be measured and compared to simulation; both signatures vary with a 30.2 year half-life for longer cooling times.
- Total neutron count rate will be measured with the PNAR detector and with the boron-tube neutron detectors in the PGET instrument and compared to simulation. This signature primarily varies with the 18.1 year half-life of ²⁴⁴Cm.
- 3. Neutron multiplication will be measured with the PNAR detector and compared to simulation; a signature that is expected to vary by ~5% as the fuel ages from 20 to 80 years [21].

Combining the above list with a total weight measurement and the 2-dimensional, pin-localizing, image of the pin gamma ray intensities produced by PGET, the challenge a 'would be' proliferator has in tricking the integrated NDA system is imposing. This proliferator would need to do all the following:

- 1. Emit gamma rays with the correct energy/energies and relative intensity from all the pins in a BWR or a VVER-440 assembly.
- 2. Emit ¹³⁷Cs photons and/or create an absolute current in the ion chambers that is consistent with the initial enrichment, burnup and cooling time of the declaration.
- 3. Produce two specific and related neutron count rates when the assembly is measured in two different neutrons reflecting setups. The relative intensity of the count rates, which is the indication of the level of multiplication, must be consistent with the initial enrichment, burnup and cooling time of the declaration.
- 4. Keep the assembly weight within the uncertainty limits of the weight measurement.

Given (a) the time varying complexity of the signatures ($T_{1/2}$ = 30.2 years for 662 keV photons from ¹³⁷Cs, $T_{1/2}$ = 18.1 years for total neutrons given the dominance of ²⁴⁴Cm, while the multiplication remains nearly constant as function of time) and (b) the pin level resolved image from PGET, the proposed NDA system is "difficult to trick with pin substitution"; hence ASTOR recommendation (e), is satisfied.

7. Conclusion

The PNAR instrument is a robust instrument made from mature off-the-shelf hardware. Combined with a PGET, the integrated instrument satisfies all the characteristics suggested by the NDA Focus Group convened by the IAEA as part of the ASTOR Experts Group: (a) For pin level detection, PGET is expected to be able to detect single and multiple missing pins in BWR and VVER-440 fuel. If there will be cases where this detection capability is not fully assured, the integrated NDA system will detect the absence of fuel when significant inconsistencies are detected among the multiplication, total neutron or gamma signatures. (b) For declaration verification, the measured multiplication, neutron and gamma signatures will be compared to the calculated values for each of these signatures that used the declaration as input values. (c) The multiplication will be measured by a PNAR instrument calibrated with known assemblies. (d) The hardware is expected to be robust and low-maintenance. (e) Given the range of measured signatures: spatial gamma ray emission, total neutron and gamma count rates, multiplication and assembly weight, combined with declaration-based analysis; the overall system is difficult to trick with pin substitution.

8. References

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