LA-UR-12-23035

Approved for public release; distribution is unlimited.

Title: Simulated Performance of the Integrated Passive Neutron Albedo Reactivity and Self-Interrogation Neutron Resonance Densitometry Detector Designed for Spent Fuel Measurement at the Fugen Reactor in Japan

Author(s): Ulrich, Timothy J. II
Lafleur, Adrienne M.
Menlove, Howard O.
Swinhoe, Martyn T.
Tobin, Stephen J.
Seya, Michio
Bolind, Alan M.

Intended for: INMM, 2012-07-15/2012-07-19 (Orlando, Florida, United States)

Simulated Performance of the Integrated Passive Neutron Albedo Reactivity and Self-Interrogation Neutron Resonance Densitometry Detector Designed for Spent Fuel Measurement at the Fugen Reactor in Japan

T.J. Ulrich, Adrienne A. LaFleur, Howard O. Menlove, Martyn T. Swinhoe and Stephen J. Tobin
Los Alamos National Laboratory, NM 87545

Michio Seya and Alan M. Bolind
Japan Atomic Energy Agency, Japan

Abstract
An integrated nondestructive assay instrument, which combined the Passive Neutron Albedo Reactivity (PNAR) and the Self-Interrogation Neutron Resonance Densitometry (SINRD) techniques, is the research focus for a collaborative effort between Los Alamos National Laboratory (LANL) and the Japanese Atomic Energy Agency as part of the Next Generation Safeguard Initiative. We will quantify the anticipated performance of this experimental system in two physical environments: (1) At LANL we will measure fresh Low Enriched Uranium (LEU) assemblies for which the average enrichment can be varied from 0.2% to 3.2% and for which Gd laced rods will be included. (2) At Fugen we will measure spent Mixed Oxide (MOX-B) and LEU spent fuel assemblies from the heavy water moderated Fugen reactor. The MOX-B assemblies will vary in burnup from ~3 GWd/tHM to ~20 GWd/tHM while the LEU assemblies (~1.9% initial enrichment) will vary from ~2 GWd/tHM to ~7 GWd/tHM. The estimated count rates will be calculated using MCNPX. These preliminary results will help the finalization of the hardware design and also serve a guide for the experiment. The hardware of the detector is expected to be fabricated in 2012 with measurements expected to take place in 2012 and 2013. This work is supported by the Next Generation Safeguards Initiative, Office of Nuclear Safeguards and Security, National Nuclear Security Administration.

Introduction

The NGSI Spent Fuel Project is a nominally 5-year effort involving six U.S. DOE Laboratories several universities and international collaborators. It began in March of 2009. The first two years focused primarily on the simulated performance of 14 individual NDA techniques[1-3]. In down selecting among a range of NDA techniques, the NGSI Spent Fuel Project has (1) put an emphasis on techniques that are robust and relatively easy to deploy, (2) increased the "quality" of the measured signal, and (3) planned integration into the research effort from the beginning. The PNAR + SINRD system has the features of being robust, inexpensive and simple to deploy, and as such was chosen by the JAEA for implementation at the decommissioned Fugen reactor for testing Pu quantification and addressing potential shipper/receiver differences between reactor sites and the Tokai reprocessing plant. A key research question is if the PNAR technique is sensitive enough. A key implementation concern is if facilities are willing to use an instrument that needs the fuel to pass through a collar with ~1 cm tolerances on all sides.

PNAR + SINRD

Both the Passive Neutron Albedo Reactivity (PNAR) and the Self Interrogation Neutron Resonance Densitometry (SINRD) techniques have been demonstrated to have the ability to quantify the fissile content in spent fuel assemblies.

PNAR

The concept of a PNAR technique is described by Menlove and Beddingfield[4] to measure the reactivity and fissile content of highly radioactive nuclear materials such as spent fuel from energy producing nuclear reactors. In their paper, Menlove and Beddingfield[4] used the passive neutron multiplicity counter (PSMC) to take measurements of neutron counts, which has since been replaced by the use of fission chambers (FC's) as was done by Conlin et al[5].

The PNAR technique uses intrinsic neutron emissions to self-interrogate the fissile material making two measurements of the fissile material: one with a cadmium liner around the material and another without cadmium. The ratio of the neutron count *without* the cadmium to the neutron count *with* the cadmium can be calculated and is called the *cadmium ratio*. Menlove and Beddingfield[4] found in their measurements that the cadmium ratio scales with the fissile content of the radioactive material. Thus the PNAR system, once calibrated, can be an effective way to measure the relative fissile content between spent fuel assemblies. This quantification of fissile content is conducted using the concept of *effective mass*[6]

$$^{239}\mathrm{Pu}_{\mathrm{eff}} = C_1 M_{U235} + M_{Pu239} + C_2 M_{Pu241}, \qquad (1)$$

where the constants C_1 and C_2 weight the contributions of ^{235}U and ^{241}Pu in the measured signal respective to the amount of ^{239}Pu.

SINRD

Similar to the PNAR technique, SINRD relies on ratios of neutron counts to quantify the fissile content in spent nuclear fuel. The neutron counters in SINRD are FC's wrapped in different materials ("bare", i.e. no wrapping material, polyethylene, Cd, Gd and Hf) in order to count neutrons in different regions of the energy spectrum. Lafleur et al [REF] found that the sum of the fissile Pu masses (M_{Pu239} + M_{Pu241}) correlates to the use of the ratio

FFM/(Gd+Hf-Cd), involving three FC's: 1) Fast Flux Monitor (FFM) FC located behind 1cm thick B_4C, 2) Gd+Hf,, FC wrapped in a layer of Gd and a layer of Hf metal foils, and 3) Cd, FC wrapped in Cd foil.

Instrument Design

To perform the measurements necessary to calculate the Pu quantification ratios of PNAR and SINRD, a detector system was designed to house 12 fission chambers. PNAR necessitates that these FC's be distributed in height so as to accommodate the ability to simultaneously perform measurements with and without a Cd shield in place around the spent fuel assembly (SFA). Additionally, the Fugen SFA's have a cylindrical symmetry, which further defines the geometry of the detector. Finally, the facility requirement to maintain a ~7 mm clearance around the SFA during the measurement finalizes the size radially, with the total height being constrained to within 59.0 cm; both requirements being dictated by the facility to match instrumentation racks in the spent fuel pool at Fugen. Figure 1 provides a schematic of the detector system as represented in MCNPX.

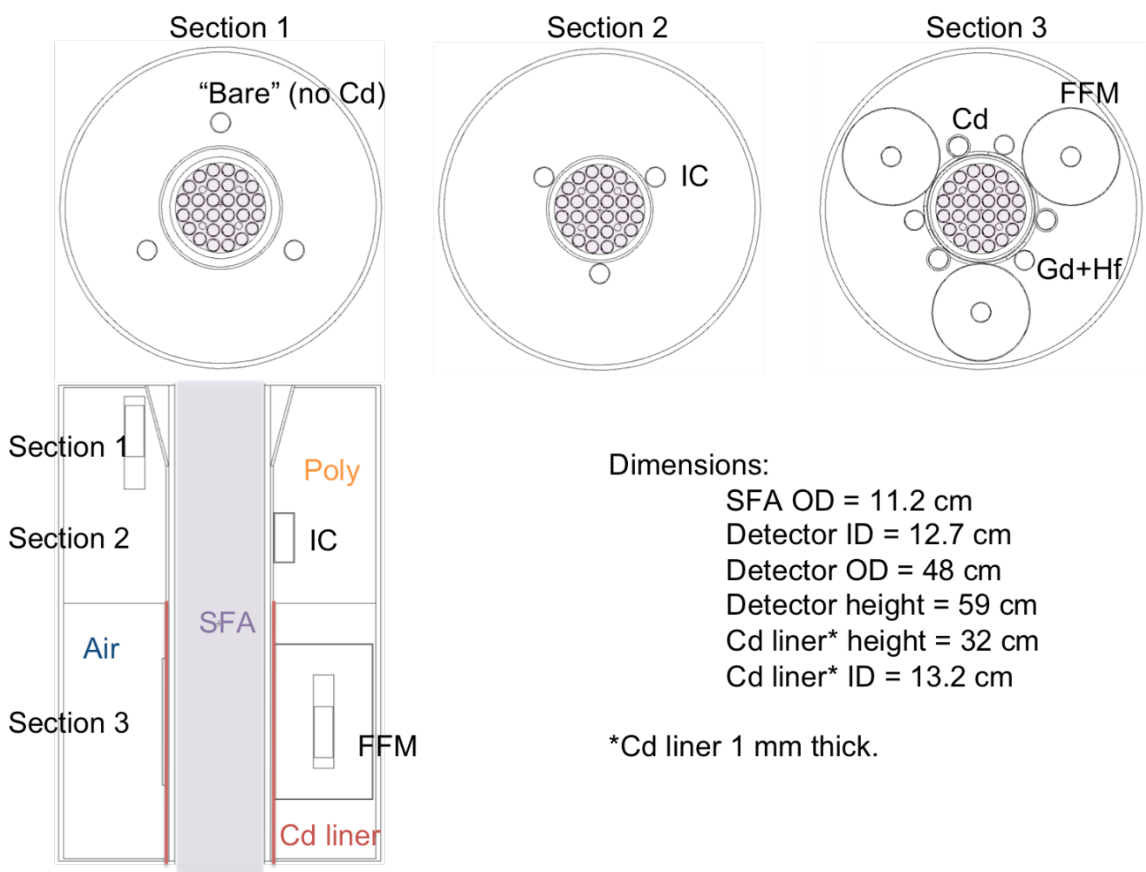

Figure 1. MCNPX representation of the PNAR+SINRD detector system for use at the Fugen site in Japan.

The FC's used in this detector system differ involve the layers of Cd, Gd and Hf in the same fashion, and nominally the same thicknesses, as the previous detector system used by Lafleur. Two exceptions to this are the lack of a true "bare" indicator, as the FC labeled “bare” in Fig. 1 is embedded in polyethylene in sec. 1 and the FFM is also embedded in polyethylene and wrapped with an addition Cd layer and the B4C4 has been removed. As a result the traditional SINRD ratio for Pu quantification is affected and a new ratio is used in its place. This new ratio is presented in the following section containing the modeling results.

The circular cross section of the Fugen SFA’s, along with the possibility of the SFA being located off-center of the detector due to the 7mm water gap surrounding the SFA, provide the motivation for using three identical FC’s for each FC type. These three FC’s are situated in a three-fold symmetry arrangement, being positioned at 120° relative to one another. A 3-D rendering made from the mechanical drawings of this detector system can be found in Fig. 2.

The PNAR system utilizes the “bare” (i.e., unwrapped) FC appearing in sec. 1 and the FFM appearing in sec. 3 (the Cd section). The active lengths of these two sets of FC’s are separated vertically to minimize the effect of the presence of the Cd shield on the “bare” FC measurement. For SINRD, all of the metal wrapped FC’s reside in sec. 3. The FFM and Cd wrapped FC’s each lay behind the Cd shield which is in place for PNAR. The Cd shield has 1” wide windows (2.5” tall to match the active length of the FC’s) removed with Gd and Hf layers added in those windows in order to create the Gd+Hf filter for the remaining three FC’s in sec. 3. Finally, total gamma measurements, not reported on in this document, are made using the ion chambers that appear in sec. 2.

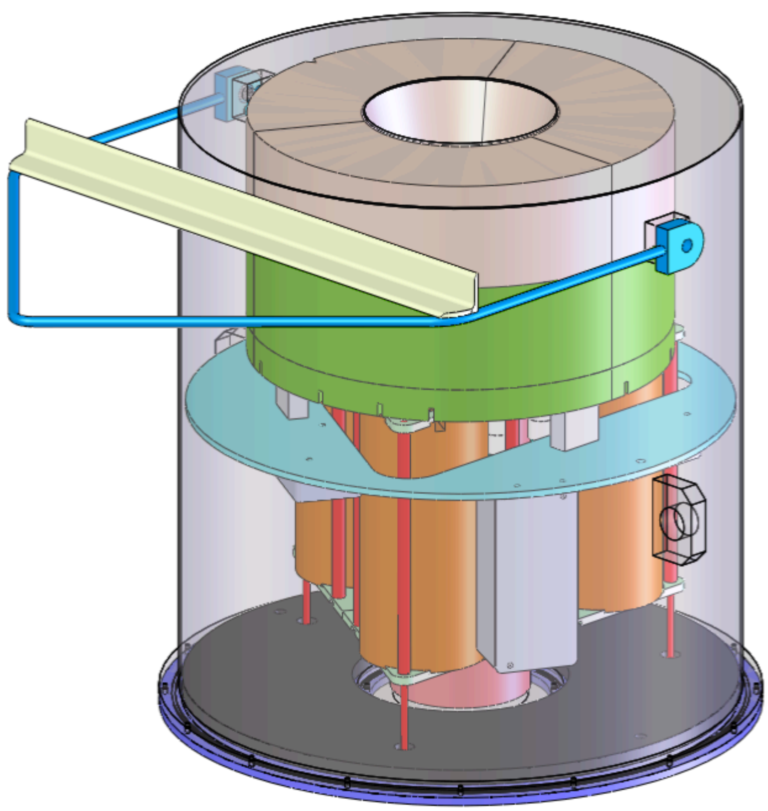

Figure 2. 3-D rendering of the internal and external structure of the detector system.

MCNPX Results

The geometry shown in Fig. 1 is taken directly from the MCNPX model. This model of the detector was used in MCNPX simulations with SFA's of five different burn-ups (BU = 3, 7, 10, 15, and 19 GWd/MTU) at a cooling time (CT) of 20 years. The BU's selected represent those available for measurement at the Fugen site during the measurement campaign planned for the summer of 2013. The results from these simulations are presented here. First, however, we must address the degree of freedom in SFA position that arises from the differing dimensions of the outer diameter of the SFA (11.2 cm) and the inner diameter of the detector (12.66 cm).

The difference in diameters mentioned above gives rise to the possibility of the nominal 7 mm water gap to reduce to 0 mm on one side while simultaneously increasing to 14 mm on the other. The three-fold symmetry of the FC layout then was implemented to mitigate differences that each individual FC response that may arise as a result. For all PNAR and SINRD ratios, therefore, the ratio is taken using the sum of the 3 identical FC's. Also, the three-fold symmetry allows us to restrict the cases for which the SFA may be misaligned to within a 60° arc. To explore the effect of the misalignment of SFA and detector ten positions were used in the simulations. These positions are illustrated in Fig. 3 and were only used for the 19 GWd/MTU cases.

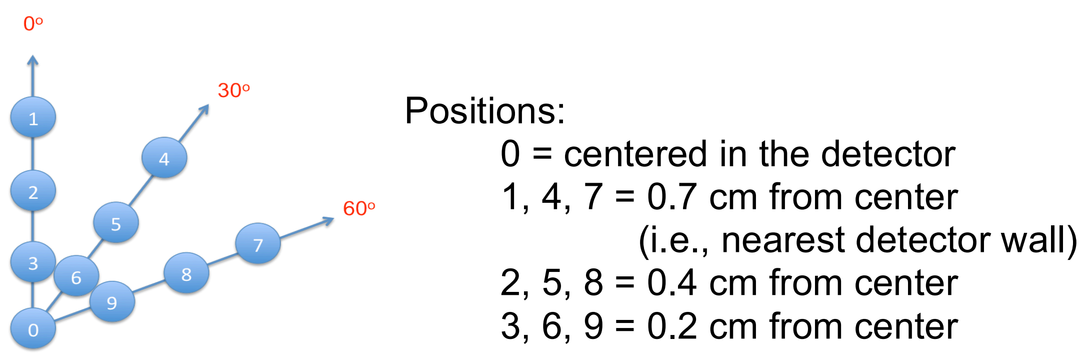

Figure 3. Variable positioning of the SFA in the detector.

The results from MCNPX simulations for the variable SFA positioning, as well as the variations due to BU and CT can be found in Figs. 4 and 5 for PNAR and SINRD respectively. The results for each measurement type are shown plotted against their respective Pu quantities for which they are known to be sensitive (i.e., $^{239}Pu_{eff}$ for PNAR and ^{239}Pu + ^{241}Pu for SINRD). For the PNAR results, the C_1 and C_2 required for the $^{239}Pu_{eff}$ calculation are 0.31 and 0.16 respectively, as taken from the MCNPX tally in the fuel. Note the change in the SINRD ratio, i.e., using the "bare" FC in place of the FFM.

Both PNAR and SINRD show similar sensitivity as measured by the slope of the ratio as a function of the mass of Pu, indicating approximately 20% change over the range of BU. Closer examination, however, indicates the positional variations of the SFA affect the SINRD ratio much more strongly than in PNAR. This fact thus indicates a greater level of uncertainty on the SINRD measurements without a method to know and compensate for the positional fluctuations.

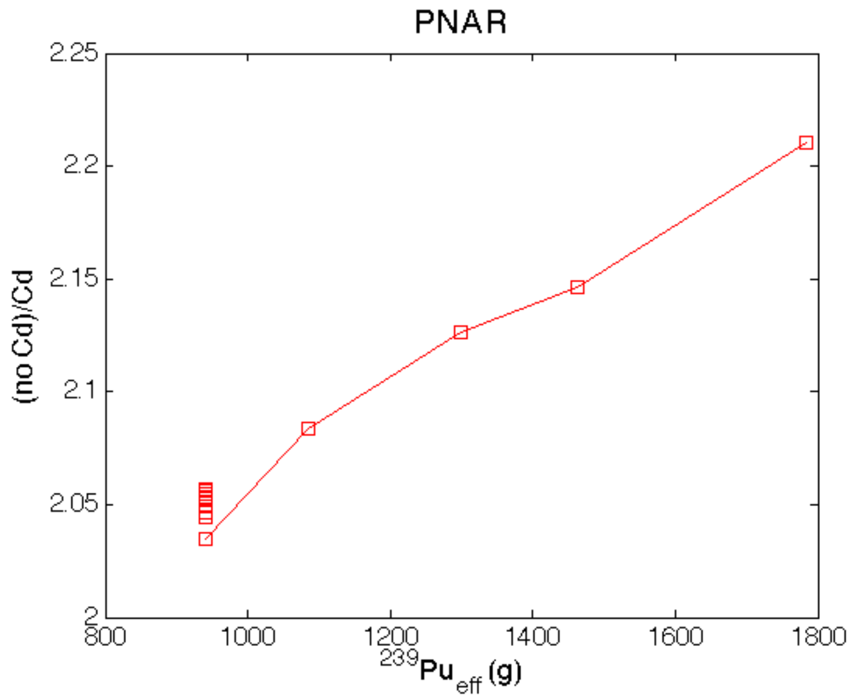

Figure 4. Pu quantification ($^{239}Pu_{eff}$) results from MCNPX simulations for PNAR.

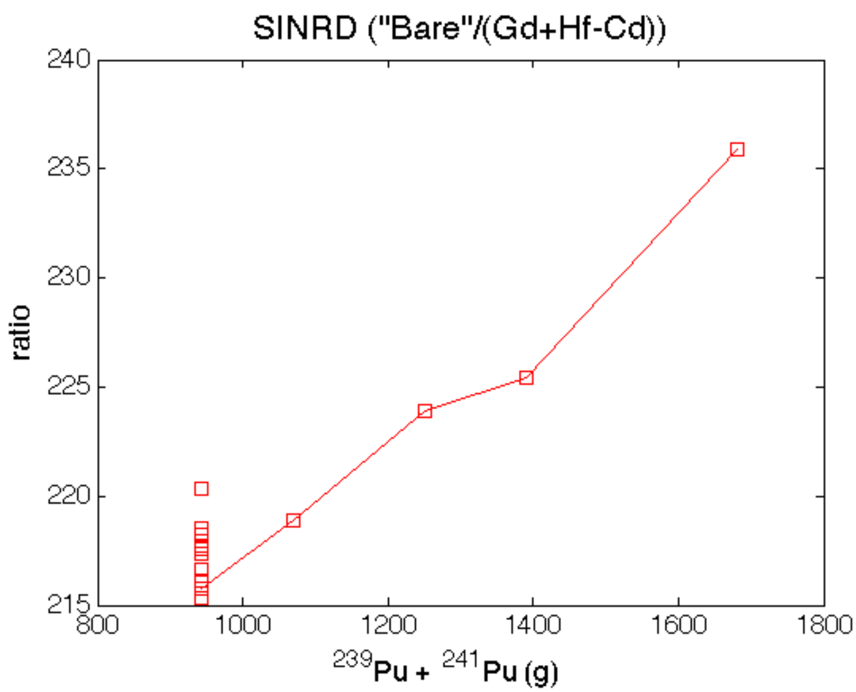

Figure 5. Pu quantification (^{239}Pu + ^{241}Pu) results from MCNPX simulations for PNAR.

Conclusion

Initial results from simulating realistic Fugen assemblies in the designed PNAR+SINRD detector show a great deal of promise despite the known issues that arise due to a large water gap between detector and SFA. The three-fold symmetry of the detector successfully minimizes the variations due to misalignment of detector and SFA that may arise in the upcoming measurements (summer 2013). As expected from recent studies8, SINRD is most affected by the water gap and the degree of freedom added due to misalignment, while PNAR is less affected. The suspected failure of the traditional SINRD ratio (i.e., FFM/(Gd+Hf-Cd)) for Pu quantification arises due to effect of the water gap and the thermalization of the neutrons emanating from the SFA as they travel to the FC's through the excess water. The water gap thus decreases "slows" the FFM such that it does not measure the fast flux in the way originally used by Lafleur. As a result the "bare" FC, which is more sensitive to the detection of thermal neutrons, proves to be necessary for SINRD to provide a monotonic correlation between the ratio and the Pu mass.

Additionally the MCNPX simulations provide an estimate of measurement times, which should be approximately 5 min. per measurement location. The measurement campaign scheduled for summer 2013 in Japan will be conducted over the course of 1 week. The relatively short measurement time will allow several SFA's with various BU's to be measured at a variety of positions, both laterally in the detector and vertically along the SFA.

Acknowledgements

This work is supported by the Next Generation Safeguards Initiative, Office of Nuclear Safeguards and Security, National Nuclear Security Administration and the Japan Atomic Energy Agency. We extend special thanks to the staff at the decommissioned Fugen reactor in Japan for their contributions to the mechanical specifications of the integrated detector system and their cooperation in accommodating the upcoming measurement campaign.

References

[1.] Veal, K.D., S.A. LaMontagne, S.J. Tobin, and L.E. Smith. 2010. NGSI Program to Investigate Techniques for the Direct Measurement of Plutonium in Spent LWR Fuels by Nondestructive Assay, Institute of Nuclear Materials Management 51st Annual Meeting, Baltimore, MD, July 11-16, 2010.

[2] Humphrey, M.A., S.J. Tobin, and K.D. Veal. 2012. The Next Generation Safeguards Initiative's Spent Fuel Nondestructive Assay Project, Journal of Nuclear Materials Management, Vol. 40, No. 3, (Spring 2012) pp. 1-6.3

[3.] Tobin, S.J., H.O. Menlove, M.T. Swinhoe and M.A. Schear, "Next Generation Safeguards Initiative research to determine the Pu mass in spent fuel assemblies: Purpose, approach, constraints, implementation, and calibration," Nucl. Instr. Meth. in Physics Research A 652, pp.73-75, 2011.

[4] Menlove, H. O., and Beddingfield, D. H. (1997) Passive Neutron Reactivity Measurement Technique.

[5] Conlin, J. L., Tobin, S. J., Hu, J., Lee, T. Menlove, H.O., and Croft, S. (2011) Passive Neutron Albedo Reactivity with Fission Chambers., *Los Alamos National Laboratory document LA-UR-11-00521.*

[6](1991) *Passive Nondestructive Assay of Nuclear Materials.*
[7]LaFleur, A. M.; Menlove, H. O.; Charlton, W. S.; and Swinhoe, M. T. (2012) Development of Self-Interrogation Neutron Resonance Densitometry to Quantify the Fissile Content in PWR Spent LEU and MOX Assemblies, *Nuclear Science and Engineering,* **171**, 1 – 17.
[8] Hu J., Trellue, H.R., Tobin, S.J., Ulrich, T., LaFleur, A.M., Freeman, C.R., Menlove, H.O. and Swinhoe, M.T., (2012) The Performance of Self-Interrogation Neutron Resonance Densitometry in Measuring Spent Fuel, *J. Nuc. Mat. Managment*.