INL/CON-09-16103
PREPRINT

Measurement and Characterization of Nuclear Material at Idaho National Laboratory

INMM Annual Meeting

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July 2009

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Abstract

A measurement plan and preliminary Monte Carlo simulations are presented for the investigation of well-defined mixed-oxide fuel pins. Measurement analysis including pulse-height distributions and time-dependent cross-correlation functions will be performed separately for neutrons and gamma rays. The utilization of Monte Carlo particle transport codes, specifically MCNP-PoliMi, is discussed in conjunction with the anticipated measurements. Four EJ-309 liquid scintillation detectors with an accurate pulse timing and digital, offline, optimized pulse-shape discrimination method will be used to prove the dependency of pulse-height distributions, cross-correlation functions, and material multiplicities upon fuel pin composition, fuel pin quantity, and detector geometry. The objective of the measurements and simulations is to identify novel methods for describing mixed-oxide fuel samples by relating measured quantities to fuel characteristics such as criticality, mass quantity, and material composition. This research has applications in nuclear safeguards and nonproliferation.

Keywords: Cross-correlation, Plutonium-oxide, Liquid scintillator, Special nuclear material, MCNP-PoliMi

1. Introduction

The need for advanced safeguards techniques to accurately characterize nuclear fuels containing plutonium and other transuranic elements is increasing in demand as the desire to utilize nuclear power as a reliable energy source increases. In this context, fuel reprocessing and advanced fuel recycling are important topics in the nuclear power industry. Mixed-oxide (MOX) fuels utilize plutonium that persists after the use of reactor fuel. Re-use of both plutonium and uranium in the form of MOX fuels offers a significant increase in the amount of total energy produced from the fuel material [1].

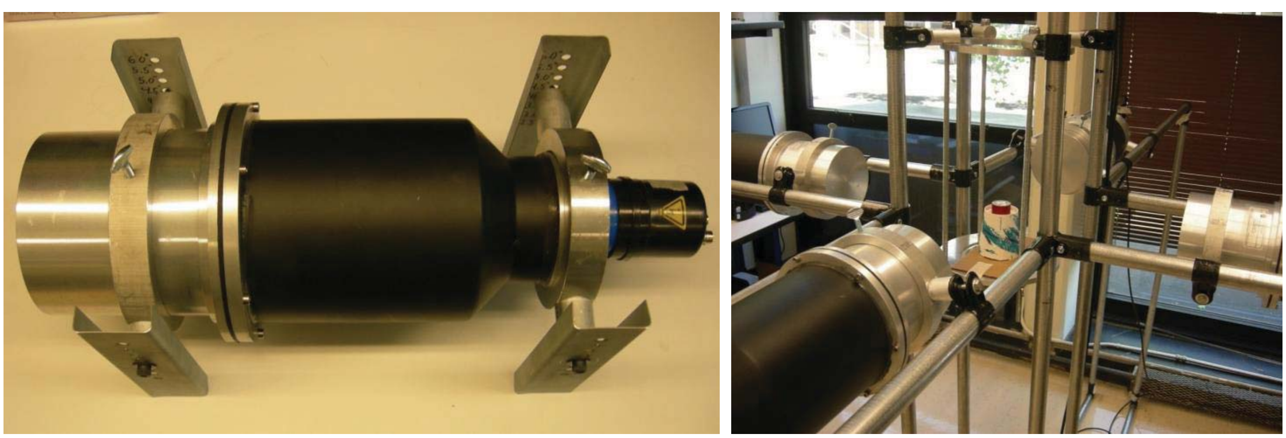
Organic scintillation detectors are being increasingly used in systems that are developed to measure both neutrons and gamma rays from fissile materials such as MOX. These detectors function at an appropriate range of energy for neutron detection within this application (typical neutron-measurement range is between 500 keV and 10 MeV), allowing high-energy neutron detection without moderation [2]. In addition to neutron detection, organic scintillators are sensitive to gamma rays. This dual mode of detection makes organic scintillators viable in applications requiring the detection and characterization of special nuclear material (SNM). Furthermore, liquid scintillators offer the capability to post-process measured data utilizing pulse-shape discrimination (PSD), thus providing an accurate method for distinguishing between neutrons and gamma rays [3]. The PSD method has been established in the past and is based on standard charge-integration method. Specifically, two integrals are calculated for each measured pulse: an integral of the pulse tail and an integral of the total pulse. The two range-optimized integrals allow the calculation of a ratio to distinguish the interacting particle type.

Recently, a measurement system developed at the University of Michigan (UM) was used to measure plutonium-oxide samples at the JRC in Ispra, Italy: pulse-height distributions (PHDs), cross-correlation functions, and multiplicities were acquired. The amplitude of the PSD-attributed neutron and gamma-ray pulses, which is a function of incident particle energy, is used as the basis for creating PHDs [4]. Cross-correlation functions are derived from differences between the arrival times of two correlated detections [5]. The Monte Carlo particle transport code, MCNP-PoliMi, has the capability to accurately model interactions necessary for these measurements [6]. This paper presents new simulation results of cross-correlations from fresh MOX fuel pins; these cross-correlations will be measured at the Idaho National Laboratory (INL) in June of 2009 and this novel measurement will result in a large amount of data that will be used to validate Monte Carlo results. The ultimate goal of this measurement is to provide new methods for the detection and characterization of MOX fuel elements that will be accurate, fast, and robust.

2. Measurement Description

A. Description of Measurement Set-up

Figure 1 shows a single EJ-309 detector, secured to its height-adjustable holder. The measurements will be performed using four EJ-309 liquid scintillation detectors. The detectors will be placed horizontally in 90° intervals around a MOX sample, with each detector equidistant from the sample (see MCNP-PoliMi model in Figure 3a). Lead bricks will surround a MOX fuel pin assembly as necessary to appropriately attenuate the fuel assembly's gamma-ray background. A CAEN V1720, 8-channel, 12-bit, 250-MHz digitizer with real-time sampling capability will be used to digitize and store measured pulses. Each of the four channels provides time-synchronized pulse information which is collected only when exceeding the applied 70 keVee (keV electron equivalent) light output threshold (corresponding to approximately 450 keV neutron deposited energy). This digital data acquisition system enables the implementation of pulse-height and time correlation algorithms enhanced by optimized offline PSD methods [3].



(a) (b)

Figure 1. a) EJ-309 liquid scintillation detector with height-adjustable stand, b) Detector geometry for cross-correlation measurements of ^{252}Cf

The detectors were calibrated to the same gain using a ^{137}Cs source. The data acquisition system was tested using a 12-μCi ^{252}Cf neutron source as well as a 1-Ci Pu-Be neutron source.

Two MOX pin types will be measured at INL jointly by UM and INL personnel. The measurements will be performed with a measurement system developed at UM to measure PHDs, cross-correlation functions, and multiplicities. The dependence of these measured quantities on fuel pin composition, fuel pin quantity, and detector geometry will be determined. The material compositions of the pins are shown in Table 1 where a notable difference can be observed between the pins in the mass of ^{240}Pu. This isotope is the strongest spontaneous-fission neutron source in the MOX pins. In addition to dependence upon fuel pin material composition, the ability to detect differences in fuel pin quantity will also be assessed. The measurements will be performed on a quantity of approximately 100 fuel pins (equivalent to approximately 1 kg of plutonium), for the two fuel types, and an additional configuration of approximately 50 fuel pins will be available for one of the fuel types. The final measurements will be performed with varying sample-detector distance.

Table 1. Isotopic compositions of two MOX fuel pin types at the INL [8].

Isotope	Pin #1 (wt. %)	Pin #2 (wt. %)
^{238}Pu	0.01	0.01
^{239}Pu	11.42	10.98
^{240}Pu	***1.53***	***4.10***
^{241}Pu	0.17	0.58
^{242}Pu	0.02	0.02
^{241}Am	0.06	0.16
^{235}U	0.17	0.16
^{238}U	74.78	72.13
O	11.85	11.86

B. Data Analysis and Expected Results

The measured data will be processed by optimized, offline, digital PSD techniques. The data acquired during each measurement configuration of the MOX fuel pin assemblies will be processed to obtain PHDs and cross-correlation functions.

The amplitude of the PSD-attributed neutron pulses is strongly related to the incident neutron energy. Despite this relationship, when using organic scintillation detectors, the resulting PHDs require the use of spectrum unfolding to obtain incident neutron energy spectra. Additionally, time of flight (TOF) measurements can be used to confirm neutron energy spectra obtained through PHDs and Monte Carlo simulations. Measured energy distributions provide insight into the presence of oxide in a mixed-oxide fuel pin assembly. For this purpose, the presence of valleys in the unfolded energy spectra, significance of (α, n) contributions, and changes in the average detected neutron energy are employed as discussed in Section 3. The average detected energy provides information with regards to the quantity of fuel pins within the assembly.

Timing information will be used to calculate cross-correlation functions and material multiplicity. Separate contributions to total cross-correlation functions (i.e. neutron-neutron, gamma-ray–neutron, etc.) are identified through PSD and provide information that is unique to the sample’s material composition, constituent activity, and structural geometry. Relationships will be formulated, connecting correlation measurements and multiplicity analysis with quantities such as material criticality, mass quantification, and sample composition.

3. Monte Carlo Analysis

A. MCNP-PoliMi Description

Many Monte Carlo simulations of nuclear processes utilize interaction physics in conjunction with stochastic particle transport. Examples are the MCNP codes. However, MCNP does not incorporate the correlated particle detection required in several SNM-characterization applications. MCNP-PoliMi is a modified version of the MCNP4C code developed in order to obtain these time-correlated quantities – specifically the correlation between neutron interactions and their consequent gamma-ray production. MCNP-PoliMi utilizes a unique event-by-event modeling technique that does not stray from physical reality by using non-analog physics, unlike MCNPX [7].

The latest version of MCNP-PoliMi, version 1.2.5, incorporates the ability of simulating all standard MCNP sources with additional custom sources. These novel sources, such as ^{240}Pu and ^{242}Pu spontaneous-fission sources include spontaneous-fission distributions with specific multiplicity distributions. Additionally, (α, n) distributions are source options for situations involving plutonium isotopes in oxides. All spontaneous-fission sources and (α, n) sources from the isotopes shown in Table 1 were modeled. Figure 2 shows the contributions of the sources to the neutron production rate.

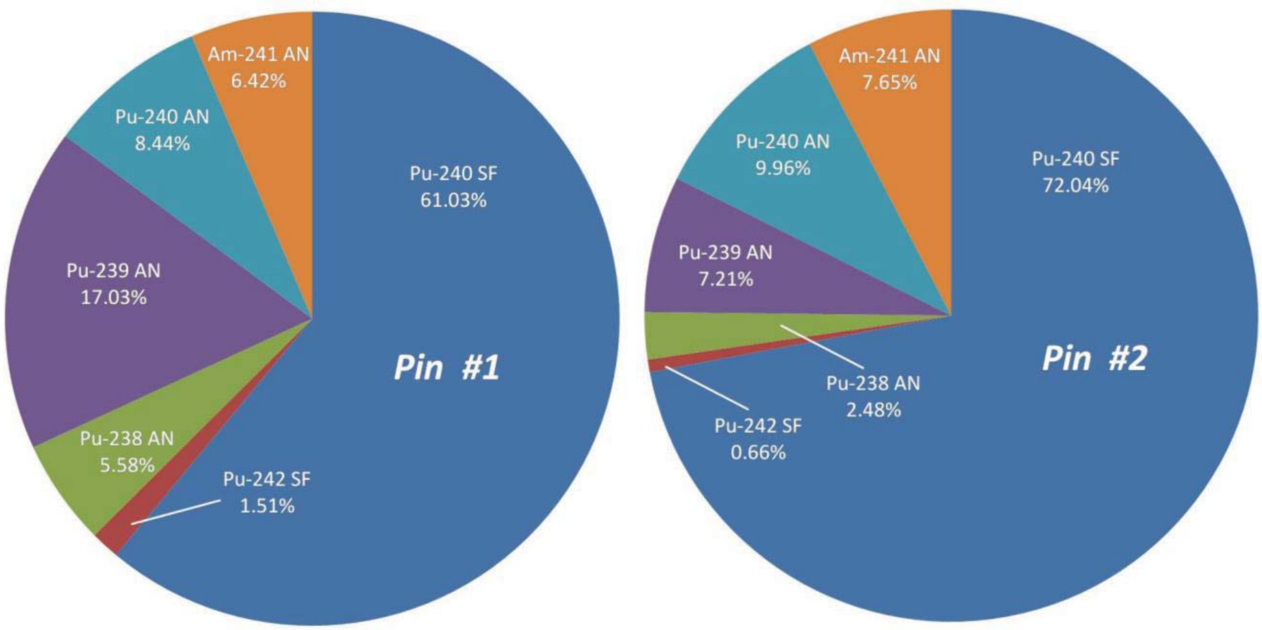
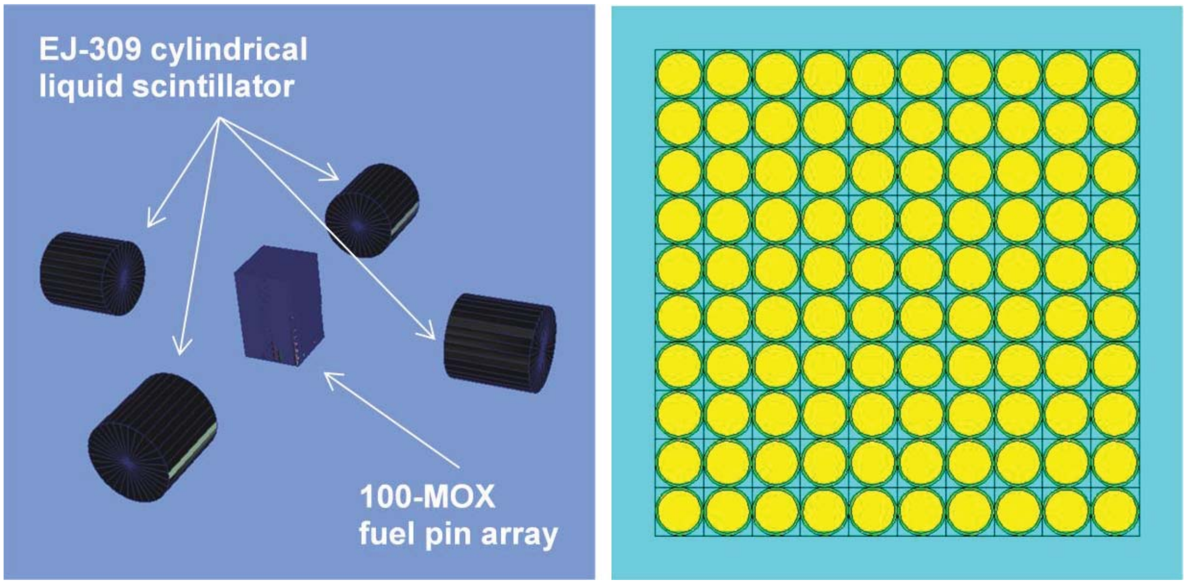


Figure 2. Contributions of spontaneous fission and (α, n) neutron sources present in the INL MOX fuel pins to the total neutron .

The data outputs from MCNP-PoliMi simulations include details about each individual interaction that took place in the detector. The data are subsequently post-processed and tailored to anticipate a particular detector's response. These simulations of well-defined INL MOX fuel pins provide the information necessary to obtain PHDs, cross-correlation functions, and multiplicities. A light-output threshold of 70 keVee is used in post-processing to discard any neutron that creates a pulse that does not produce enough light to be detected in a practical situation.

B. Description of Monte Carlo Models

The MCNP-PoliMi model of the proposed measurement set-up includes four EJ-309 liquid scintillation detectors which are placed around the axis of the MOX fuel pin set-up, with each detector equidistant from the source, as shown in Figure 3a. Parameters that are adjusted during the simulations include the composition of the fuel pins (two pin types), the distance between the detectors (30 cm and 60 cm) and the number of pins under investigation (50 and 100 pins).



(a) (b)

Figure 3. a) Three-dimensional MCNP-PoliMi model including four EJ-309 cylindrical liquid scintillators 30 cm from the axis of the fuel pin array, b) Two-dimensional depiction of the MCNP-PoliMi model of the centrally located array of 100 MOX fuel pins where yellow represents fuel and green zirconium cladding.

All measurements scenarios were simulated with the MCNP-PoliMi code in order to determine dependence of PHDs, cross-correlation functions, and multiplicities upon sample type. The sources simulated in MCNP-PoliMi were two varieties of MOX fuel pins with different isotopic compositions, as outlined in Table 1 and Figure 2 [9]. The neutrons and gamma rays emitted from each pin originate from the individual spontaneous fissions seen in plutonium or (α, n) reactions occurring in the presence of oxygen. All six source contributions in Figure 2 were modeled individually. The reported results are then the summation of the products of the six simulation results with their reaction probabilities. Exact fuel pin geometry and cladding material are specified in the literature and included into the model [8].

C. Simulation Results

Initial simulation results included the neutron energy distributions for various numbers of pins and the detector response: PHDs and cross-correlation functions.

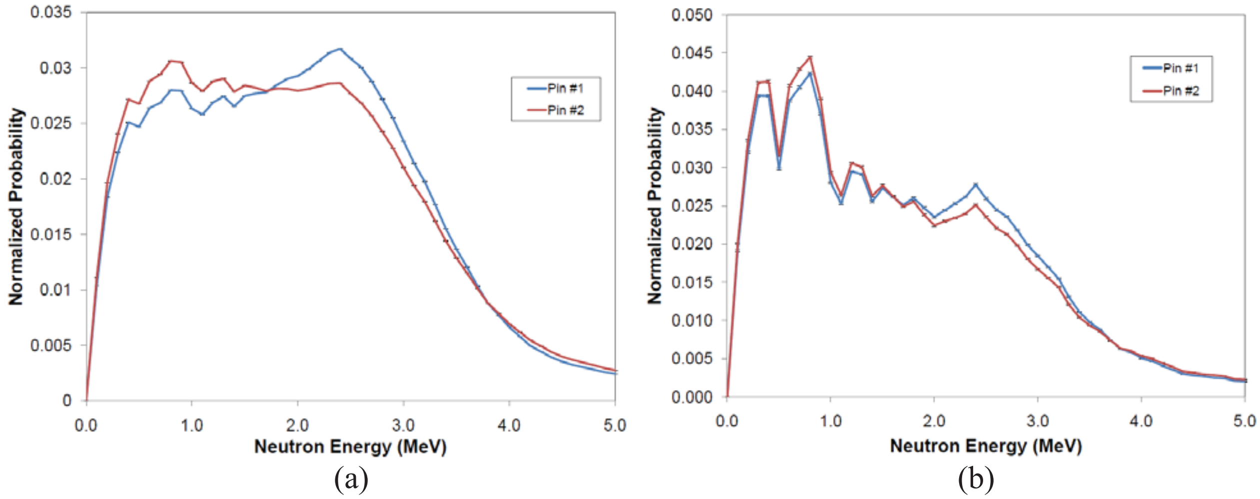


Figure 4. a) MCNP-PoliMi simulated neutron energy spectra incident on the face of the detector for a single MOX fuel pin, b) MCNP-PoliMi simulated neutron energy spectra incident on the face of the detector for a 100-pin MOX fuel assembly.

Figure 4a shows the energy distributions of the neutrons incident upon the detector from an individual MOX fuel pin for both pin types. Figure 4b shows the neutron energy distributions for 100-pin assemblies. Self-shielding and multiplication effects significantly alter the general shape of the neutron energy spectra in comparison to the single pin case. The valleys located in the lower energy region of the spectra, specifically near 0.05 MeV, 0.1 MeV, and 1.3 MeV, are due to energy resonances in the neutron elastic scattering cross-section for ^{16}O, which is shown in Figure 5 [10]. Simulations were also performed to determine that neutron energy distributions are a strong function of pin quantities with respect to both the shape of the spectra (Figure 6a) and average energy values (Figure 6b).

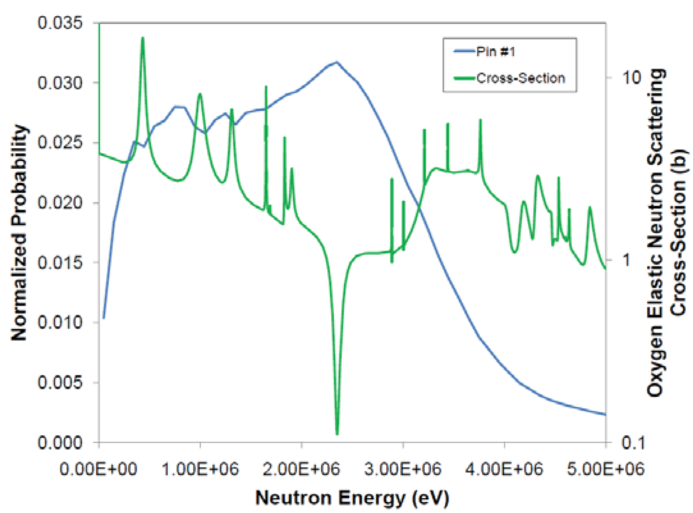
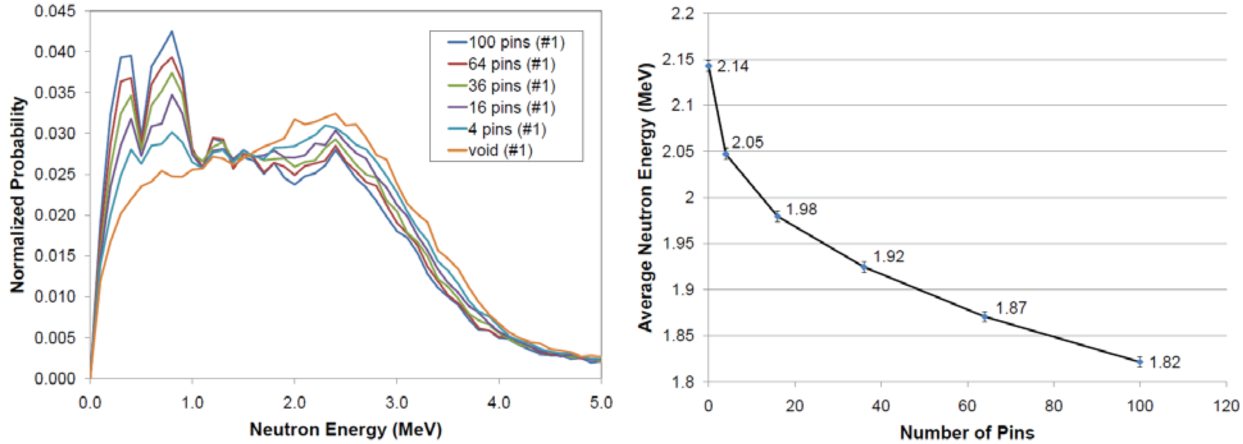


Figure 5. Comparison of the MCNP-PoliMi simulated neutron energy spectrum tallied on the face of the detector for a single MOX fuel pin type #1 and the elastic neutron scattering cross-section on ^{16}O.



(a) (b)

Figure 6. Neutron energy spectrum (a) and average detected neutron energy (b) as a function of the quantity of fuel pins (type #1) included in the assembly.

Time-dependent cross-correlation functions were generated from the simulation data output with a MCNP-PoliMi post-processor. Total cross-correlation functions for assemblies of both pin type #1 and #2 are shown in Figure 7. For correlations detected with time differences of -40 ns to 40 ns, it is anticipated that a 100-pin assembly of type #1 will produce approximately 18 cross-correlations per second (ccs/s); and a 100-pin assembly of type #2 will produce approximately 47 ccs/s. The shape of the curves are quite similar for the pin type comparison in Figure 7, although the relative amount of ccs/s varies significantly due to pin type #2's larger abundance of ^{240}Pu.

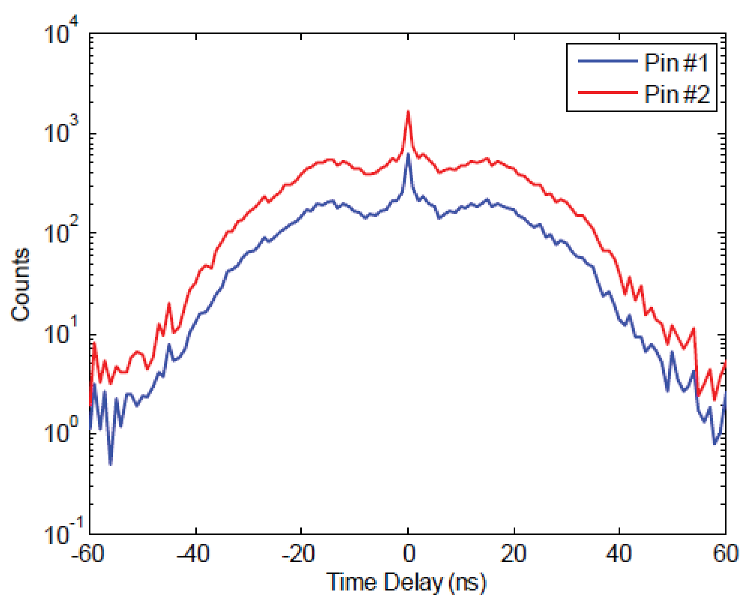
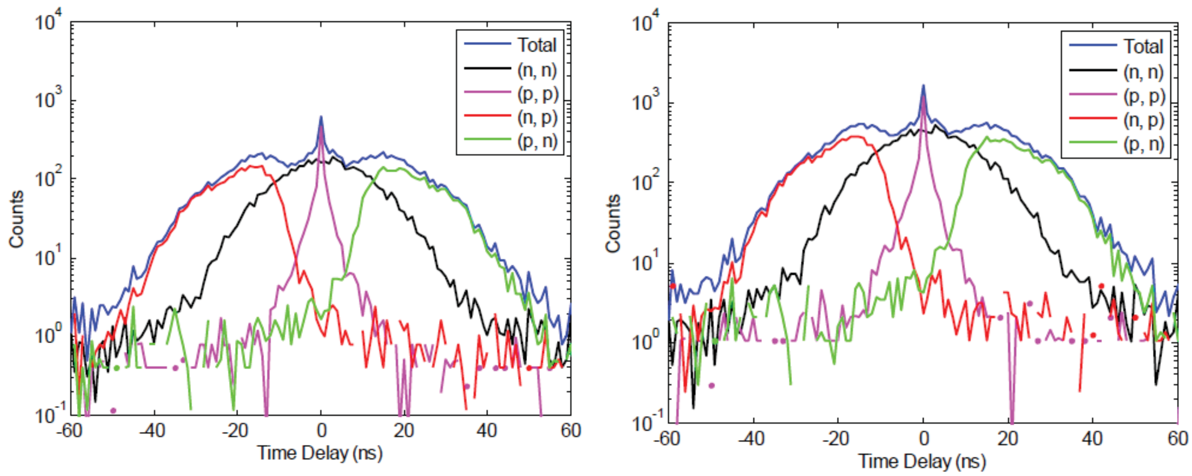


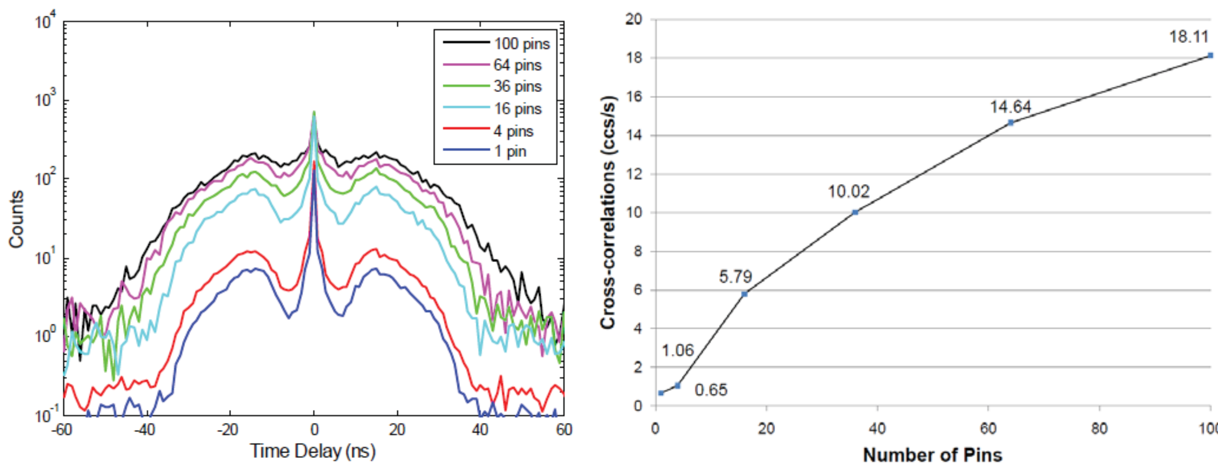
Figure 7. Comparison of the MCNP-PoliMi simulated total cross-correlation functions for pins #1 and #2 in a 10-minute acquisition time.

Each total cross-correlation curve from Figure 7 can be broken down into individual contributions as shown in Figure 8. Specific correlation curves such as the neutron-neutron curve can be used to study concepts such as multiplicity and criticality. The total cross-correlation functions as a function of pin quantity are shown in Figure 9a. Integrating these total correlation curves provides valuable information on parameters such as mass quantification; Figure 9b shows the relationship between pin quantity and the number of cross-correlations per second.



(a) (b)

Figure 8. Breakdown view of cross-correlation functions showing the individual correlation contributions for pins #1(a) and #2 (b). The simulations were normalized to a 10-minute acquisition time.



(a) (b)

Figure 9. a) MCNP-PoliMi simulated total cross-correlation functions for different pin quantities for a 10-minute acquisition time, b) trend in total cross-correlations per second for different pin quantities.

The presented correlations result from a combination of six simulations, originating from spontaneous fission and (α, n) reactions that are present in MOX fuel. Pin type #1 acquires approximately 90% of its correlations from the spontaneous fission sources and 10% as a result of the (α, n) sources. Pin type #2 acquires approximately 93% of its correlations from the spontaneous fission sources and 7% as a result of the (α, n) sources. This is expected because pin type #2 contains more ^{240}Pu than pin type #1. The passive detection of oxygen in plutonium is made possible by the (α, n) reactions which alter the shape of the cross-correlation functions, primarily the neutron-gamma and gamma-neutron components of the correlation curves.

4. Conclusions

This paper presented new simulation results for MOX fuel pin assemblies that will be measured at the Idaho National Laboratory in June, 2009. The experimental set-ups were derived from detailed Monte Carlo modeling which incorporated accurate detector response functions. We presented a detailed model of the neutron source from the MOX fuel, which includes spontaneous fission and (α, n) contributions. The neutron energy spectrum incident on the detectors for a varying number of fuel pins was determined. Neutron and gamma-ray cross-correlation functions were simulated for a varying number of pins. The separate contributions to these correlation functions were discussed and analyzed. The results show that this type of measurement can be used to distinguish the two pin types and to determine the mass (or number of pins) of the assembly. In addition to developing an experimental methodology, this study will be used as a basis for the future validation of the MCNP-PoliMi code for modeling MOX type fuel assemblies.

Future work will consider a more customized methodology in experimentally distinguishing MOX fuels. Relation between measured pulse-height distributions and cross-correlation functions, and quantities such as material criticality, mass quantification, and sample composition will be assessed in detail.

Acknowledgements

This work was supported by the U.S. Department of Energy Office of Nuclear Energy and the Advanced Fuel Cycle Initiative Safeguards Campaign. Idaho National Laboratory is operated for the U.S. Department of Energy by Battelle Energy Alliance under DOE contract DE-AC07-05-ID14517.

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