Fast Neutron Collar (FNCL) Instrument Advancements

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Abstract

A multi-disciplinary research team at Lawrence Livermore National Laboratory (LLNL) is investigating advanced hardware components, signal processing and analysis methodologies that could substantially improve operational capabilities of the CAEN SyS VeryFuel Fast Neutron Collar (FNCL) instrument for safeguards applications. This effort is addressing the following two components of the baseline FNCL instrument:

- Hardware advancement focused on fast neutron detector array assemblies, spanning a range of design options, including the choice of scintillation material, individual cell size, light readout, shielding, and moderation for improved neutron and gamma-ray efficiency, pulse-shape discrimination, pile-up rejection, and cross-talk reduction capabilities.
- Signal processing and data analysis tools for waveform characterization and neutron correlation evaluations, that could provide a self-contained estimate of the fissile material loading without relying on calibration curves and potentially eliminating an active interrogation source.

The objective for developing hardware and software alternatives is to further extend FNCL instrument capabilities and implement a robust combination of effective detection and pulse processing techniques with analysis methodology that will not require a priori empirical calibrations for a specific type of unirradiated fuel assemblies. All hardware and software modifications have been evaluated as add-ons to the baseline FNCL unit currently available at LLNL. The newly designed detector array panels will operate with the existing data acquisition hardware. The data acquisition and analysis scripts will be executed on the FNCL control computer in parallel to the current manufacturer-supplied software.

The hardware and analysis approaches developed in this effort are evaluated in comparison with the performance of the baseline FNCL instrument in laboratory experiments in a passive regime and with a range of neutron interrogation sources, using test objects with a varying amount of uranium. Critical performance parameters were determined to quantitatively characterize improvements in the FNCL operational capabilities for each new hardware solution and components of the analysis workflow.

This paper outlines the current status of the hardware, signal processing and the development of analysis methods and will present interim results from laboratory experimental demonstrations.

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Introduction

Neutron correlation analysis is an accepted non-destructive measurement technique for nuclear material characterization in safeguards applications. It enables mass determination for fissile and fissionable content in complex mixtures and in shielded configurations. When combined with other non-destructive assay signatures, typically gamma-ray spectroscopy, neutron correlations provide a comprehensive evaluation of the isotopic composition and content. A prominent implementation of this approach with the Uranium Neutron Collar (UNCL) instrument [1] has become a routine method for verification of U-235 content in fresh (unirradiated) nuclear fuel assemblies (FFAs).

The conventional UNCL instrument relies on a neutron coincidence counting [2] variation of correlation analysis by thermalizing and capturing neutrons emanating from FFAs in arrays of moderated He-3 detectors. Passive measurements of FFA neutron emissions are often accompanied by an activation measurement with an external moderated AmLi source. In this configuration, the low-energy interrogating neutrons induce fissions primarily in U-235 resulting in the emission of correlated fission neutrons that are further amplified through FFA multiplication and are recorded as a coincidence signal within a pre-defined time gate by the UNCL detector arrays. The recorded rate of double coincidences is converted into a U-235 linear density (g/cm) metric of the FFA fissile material loading using an empirically defined calibration curve.

The UNCL method has been traditionally used for verification of conventional low-enriched uranium fuels. However, it became unreliable for modern assembly loadings with higher enrichments and burnable neutron poisons, such as gadolinium oxide (Gd_2O_3). Thermal neutron captures on Gd, further complicated by its variable content and non-uniform spatial distribution have introduced significant deviations of measured coincidence rates from empirical calibrations with assay errors exceeding 10% target value stipulated in [3]. Several approaches to this problem have been investigated, including applying simulation-informed Gd correction factor or introducing Cd thermal neutron filters, but may have not offered a universal confidence or required considerable modifications to measurement times and UNCL hardware [4-7].

To address the challenges of determining U-235 material loading in modern fuel assemblies with increased enrichments and burnable poisons, the IAEA in partnership with CAEN SpA has developed a new Fast Neutron Coincidence Collar (FNCL) instrument [8]. FNCL relies on the same operational principle as UNCL, except it utilizes proton recoil scintillation detectors to determine fast neutron coincidence rates. Without the neutron thermalization period in the acquisition process, FNCL offers considerably increased time resolution for registering fission neutron coincidences, reducing random contributions and overcoming neutron poison effects. At the same time, fast neutron detector arrays implemented with EJ-309 liquid scintillators require significantly complex pulse processing and data acquisition electronics to enable neutron and gamma-ray particle filtering with pulse shape discrimination (PSD). A prototype FNCL instrument operated by the IAEA, has been successfully deployed at some fuel fabrication facilities and demonstrated a quick and robust capability to evaluate U-235 linear density in FFAs with varying assembly geometry, fissile material loading, and burnable poison content [9].

The implementation of large fast neutron detector arrays (as in the current FNCL prototype) for field measurements presents several operational and instrumentation challenges. Careful mitigation of fast neutron scintillators susceptibility to gamma-rays is essential to ensure that pulse shape analysis, particle discrimination, pile-up rejection, crosstalk, dead time corrections, and other effects are appropriately handled by automated procedures. The physical dimensions of photomultiplier tubes (PMTs) for the scintillation light readout limit the options for compact configurations of detector panels and impose additional constraints on the layout of shielding and moderating materials in detector enclosures. The complex data acquisition and signal processing hardware of the FNCL instrument provides opportunities to deploy new and effective analysis algorithms and methods. The existing hardware offers a versatile platform for more robust signal processing with near real-time waveform analysis and particle filtering. While the current prototype still relies on neutron coincidences (doubles) rate to determine FFA fissile content using empirical calibrations, it is equipped with sufficient processing capabilities to support more complex data evaluations. A self-contained analysis approach that would determine the U-235 linear mass without prior calibration is desirable. In addition, replacement of the AmLi interrogating neutron source with a portable DD neutron generator may significantly improve field operations.

A research team at Lawrence Livermore National Laboratory (LLNL) is currently investigating possible advancements to the FNCL instrument concept by addressing its hardware, signal processing and analysis components. This effort is informed by an extensive resident expertise in implementing fast neutron correlation analysis approaches in other nuclear threat reduction areas, such as warhead verification, radiological source characterization, and nuclear material smuggling. A detailed modeling capability of the FNCL instrument responses has been developed for this effort in two parallel simulation frameworks using MCNP and Geant4 transport codes. To facilitate this experimental and modeling investigation, the team operates an FNCL instrument prototype.

Detector Hardware Investigations

The primary focus of the hardware advancement effort is to develop the next iteration of the FNCL detector panel design that would optimize the efficiency of neutron detection, gamma-ray rejection, suppressing pile-up and crosstalk between individual cells, while reducing the size and weight of the arrays. The following optimization options were identified relative to the baseline FNCL detector setup (4x4x4-inch EJ-309 liquid scintillator with a PMT photodetector):

- *Fast neutron scintillation detector material:* EJ-309 liquid, Stilbene organic crystal, Organic Glass Scintillator (OGS), EJ-276D plastic.
- *Detector cell packaging:* individual cell size varying a range of front face dimensions and thickness, number of cells in an array, packaging and shielding.
- *Photodetection light readouts:* PMTs and Silicon Photomultipliers (SiPMs), their relative particle discrimination performance and signal processing electronics capabilities (i.e. peak shapes, noise and timing parameters).

To inform the FNCL detector panel design options with higher segmentation of detector cells, the LLNL team carried out an extended experimental evaluation of fast neutron scintillation materials and photodetector combinations. Four EJ-309 cells with the following dimensions were fabricated: 2.7 x 2.7 x 3-inch, 2.7 x 2.7 x 4-inch, 4 x 2 x 3-inch, 4 x 2 x 4-inch. In addition, Stilbene and OGS cells have been produced in the baseline 4 x 4 x 4-inch size and in smaller volume geometries (~250 ml) as suggested by modeling evaluations and prior experiences. The stilbene was cut from the existing stock grown at LLNL. The OGS-100 3% plasticizer glass detector was purchased from Blueshift Optics. The EJ-309 liquid scintillator cell was manufactured by Eljen Technology. Example photographs of individual cells from this study are shown in Figure 1. The EJ-276D cells from Eljen Technology became commercially available later in the effort and are currently being evaluated.

Figure 1. Examples of newly fabricated fast neutron scintillator cells for the optimized detector array investigations: EJ-309 liquid scintillator (various dimensions), Stilbene, OGS cells in smaller volumes (~250ml) for the high cell segmentation design concept.

The cells with the 2.7 x 2.7-inch front face dimensions were selected to populate a FNCL panel design with nine detector cells arranged in a 3-row x 3-column pattern instead of the current 2-row x 2-column array in each FNCL panel. Such a configuration may offer an improved PSD, pile-up and double-scattering rejection for the same neutron efficiency, but imposes a constraint on the light readout and can be instrumented by a single 2 x 2-inch SiPM for each cell. The cells with the 4 x 2-inch front face dimensions were selected for a FNCL panel design with eight detectors in a 4-row x 2-column pattern. This geometry allows flexibility with the choice of a light readout, and will be evaluated with both PMTs and SiPMs.

Each combination of a detector material, size and photodetection light readout was first evaluated in a benchtop experimental setup with gamma-ray and Cf-252 neutron sources. Figure-of-Merit (FOM) metric for particle discrimination performance was applied as the performance evaluation criterion for the PMT and SiPM light readout options. This metric is obtained by integrating the resulting neutron and gamma-ray populations in the PSD plot over a deposited energy threshold. By scanning over this energy threshold, FOM curves can be obtained. All FOM curves were calibrated for electron-equivalent energy using Am-241, Cs-137, and Na-22 sources (60, 511, 662, and 1275 keV). The energy deposition distributions for these gamma-ray energies were modeled with Geant4 transport code to determine their quenched energy factors relative to the Compton edge. Several voltage bias values were applied for each combination of the detector cell and light readout. Primary observations from the benchtop evaluations were:

- Stilbene material consistently outperforms an equivalent EJ-309 and OGS in the particle discrimination capability with both PMT and SiPM light readouts. However, its cost and commercial supply capacity are most probably prohibitive for large detector arrays.
- Performance of OGS is comparable to the EJ-309 in the same cell-photodetector configuration.
- PMT and SiPM scintillation light readouts demonstrate equivalent performance under nominal measurement conditions (limited count rate) and regardless of the scintillation detector material type.
- SiPM photodetection light readouts offer significant reduction in cell dimensions. However, tiling SiPM arrays to cover a larger area results in increased electronic noise and may require modified readout electronics.
- The option of a "high detector cell segmentation" array, with an increased number of smaller cells, is promising an improved fast neutron correlation counting performance, reduced

crosstalk and pile-up when compared to the “small number of larger cells” configuration (with the same total neutron efficiency).

- The 3-inch thickness of individual cells may be sufficient and more appropriate for FNCL applications when compared to the 4-inch thickness of the existing instrument.

The above findings illustrate the complex interplay between the detector panel optimization parameters. Therefore, their combined effect and cost-performance impact should be further investigated in the specific context of the FNCL instrument configuration and the expected FFA measurement conditions. To facilitate this objective, the LLNL team constructed two new “intermediate” detector panels that implemented several promising detector cell configurations. Both panels were assembled inside an empty FNCL detector housing acquired from CAEN SpA specifically for this purpose. For simplified power supply arrangements, one housing was populated with PMT-equipped cells, and the other with SiPM photodetection light readouts as illustrated in Figure 2. The resulting detector arrays are operated by the data acquisition electronics of the FNCL instrument. Experimental performance evaluations of individual cells in the intermediate panels in comparison with the baseline FNCL detectors are currently underway at LLNL.

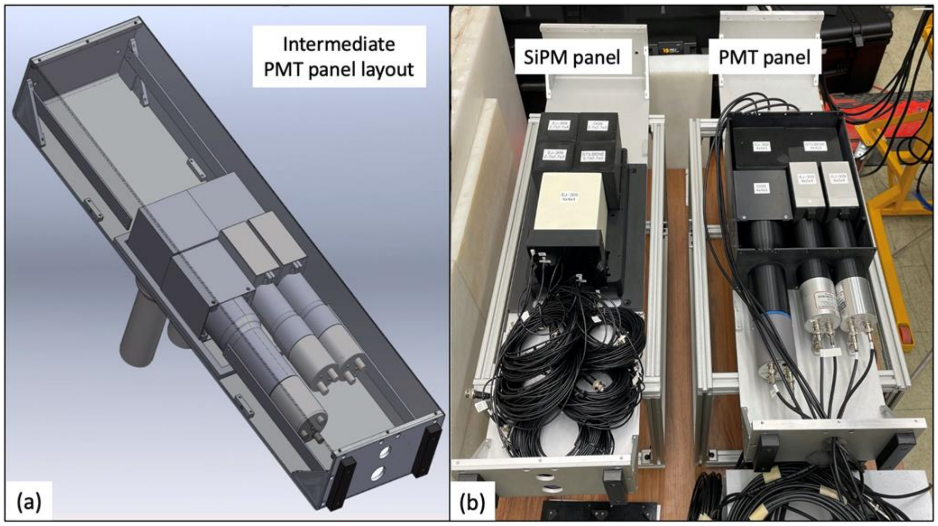

Figure 2. Two “intermediate” panels with candidate cell configurations assembled for experimental investigation with the FNCL instrument: a) the initial computer rendering of the PMT panel layout, b) fully populated SiPM and PMT intermediate panels.

Waveform Processing Advancements

LLNL is investigating an improved approach to the particle discrimination for the fast neutron detector pulses from the FNCL detector cells. The primary focus of this effort is to implement waveform processing with the Gaussian Mixture Model (GMM) technique [10,11] to perform neutron identification in a high gamma-ray background environment. In the native FNCL software, digitized waveforms from each cell are processed by applying a series of "filters” (coincidence, energy, PSD, pile-up) as specified in [8], after which the time-stamped events are recorded as a list mode file for reduction to neutron double coincidences. The GMM approach intercepts this process at the PSD step by applying neutron and gamma-ray waveform templates and a shared covariance that were determined for each detector by “training” the algorithm with the Cf-252 calibration source. In this approach, all samples are directly used to produce a z-score normalized Anscombe transformed pulse that is used for training or scoring.

An improved particle discrimination and pileup rejection capability provided by the GMM method when compared to the conventional PSD technique, is illustrated in Figure 3. In this experiment, pulses were collected with an FNCL-equivalent benchtop detector cell assembled with a 4 x 4 x 4-inch EJ-309 liquid scintillator and a PMT light readout. This cell was exposed with a Cf-252 neutron source and simultaneously with Am-241, Th-232, Na-22, Co-60, and Cs-137 gamma-ray check sources causing a considerable pileup. The same data stream was then processed with the PSD technique with the corresponding plot shown in Figure 3 (left), and the GMM method with its plot in Figure 3 (right). The GMM data processing offers a significantly improved gamma-ray and neutron discrimination as well as pileup isolation.

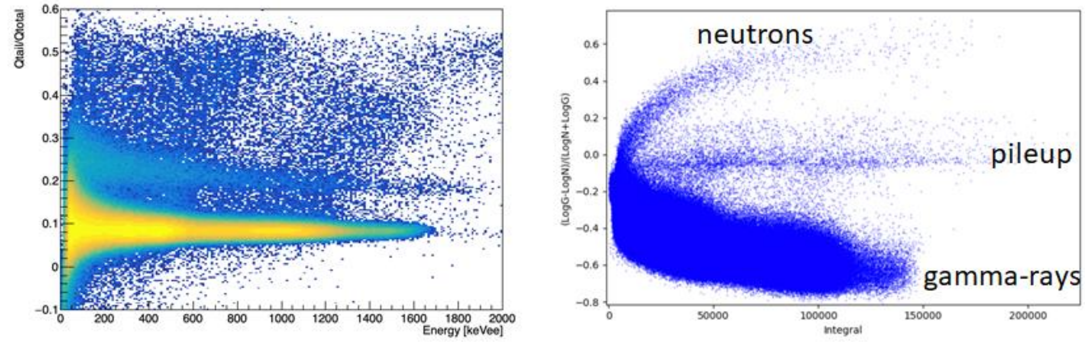

Figure 3. Comparison of the native FNCL PSD technique (left) and GMM method (right) performance for gamma-ray and neutron discrimination, pileup rejection for an FNCL-equivalent detector cell exposed with a Cf-252 neutron source with a broad-energy gamma-ray contribution from Am-241, Th-232, Na-22, Co-60, and Cs-137 gamma-ray sources.

Given the difficulty of using a truly orthogonal particle identification metric, such as time-of-flight, to evaluate GMM performance on a pulse-by-pulse basis, the LLNL team evaluated performance from score distributions in experimentally simulated gamma-ray dominated, neutron dominated, and combined measurements. The FNCL data acquisition electronics and the onboard software were used for these experimental studies by connecting a benchtop equivalent detector to a signal digitizer.

GMM-based waveform processing has been successfully implemented within an end-to-end experimental data handling and analysis workflow on the FNCL instrument. A toolkit of scripts was deployed on the FNCL computer to process digitized waveforms recorded in .bin files using user-selectable GMM or PSD particle discrimination methods. The resulting output generates neutron time history files that can be further processed with advanced analysis algorithms or reduced to coincidence count rates (i.e. rate of doubles). An illustration of this functionality is demonstrated in Figure 4. This figure shows PSD and GMM particle discrimination plots for each of the 12 FNCL detector cells with an unshielded Cf-252 neutron source inside the instrument. The same waveforms were processed with both methods using scripts executed directly on the FNCL data acquisition computer.

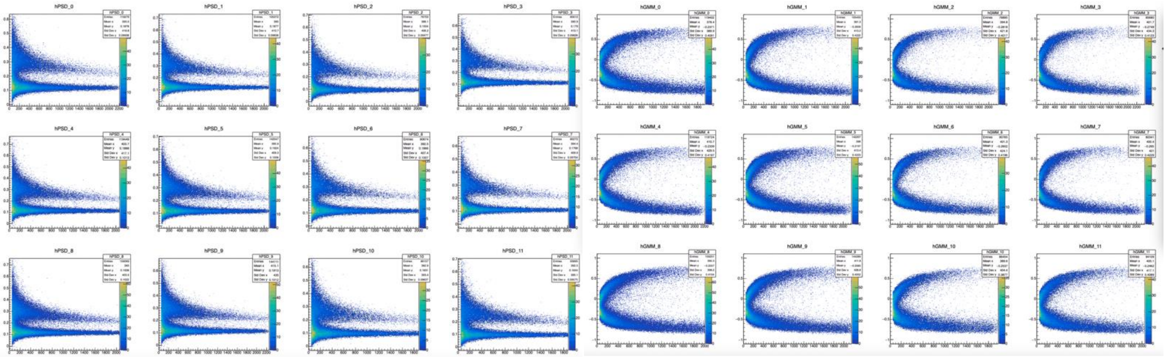

Figure 4. Illustration of the waveform processing capability implemented on the FNCL data acquisition computer. The same dataset obtained from an unshielded Cf-252 source, for all 12 FNCL detector cells, was processed with a PSD-based (top) and GMM-based (bottom) method.

The GMM-enabled FNCL data analysis workflow was used to reduce IAEA-supplied field data from field measurements of five real fuel assemblies in passive and active interrogation regimes labeled FFA_1,2,3,4, and 5. An accompanying calibration dataset was supplied to the automated GMM training routine. For this analysis, it was assumed that a pair of events scored as neutrons must be within 70 ns to be considered a double. The coincidence must also be separated by at least 14 ns if within the same panel to be considered a double; this reduces the fake double contribution from scattering. Figure 5 shows scaling between the doubles rates produced in this workflow relative to the declared U-235 linear density and the burnable poison content.

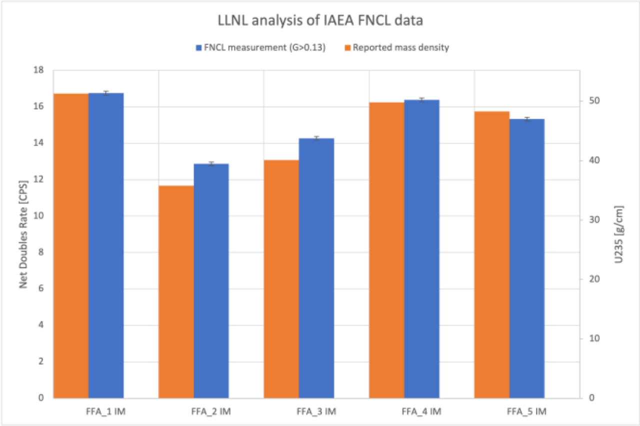

Figure 5. Doubles rates obtained using the GMM-based FNCL waveform data processing from the IAEA-supplied fresh fuel data. FFA_4 and FFA_5 contain 5.13 and 10.3 g/cm Gd_2O_3.

Self-contained analysis algorithm

The FNCL instrument capability to produce neutron time event data files motivated investigations of self-contained analysis methods for the U-235 content determination that would not require an empirical response calibration. To this end, the LLNL team has developed a likelihood fitting function for the FNCL neutron time event files based on the provisions of the fission chain restart theory. This theory [12] stems directly from the Feynman time-dependent fission chain theory [13], except that the fission chain restart theory considers two energy groups—fast and thermal neutrons—and takes into account fast neutrons thermalizing in a nearby moderator, re-entering the nuclear material, and restarting the fission chain. Two other effects that are important for the problem of assaying a fresh nuclear fuel assembly: prompt neutron escape, and fast neutrons scattering between adjacent liquid scintillator cells resulting in double counting.

The LLNL approach to the FNCL data analysis evaluates the "neutron waiting time distribution" (time between two consecutive neutron detection events) and is based on a previously developed sequential Bayesian sampling-importance-resampling particle filter technique for processing of the neutron data. The Online Statistical Analysis of Neutron Time Correlations (OSANTC) algorithm [14-16] used a likelihood function based on the Böhnel theory [17] which is the t⟶∞ limit of Feynman's time-dependent fission chain theory.

A neutron waiting time distribution of any multiplying system has several generic features illustrated by a Geant4 modeling of the FNCL response shown in Figure 6. This distribution is fitted with the theory-based likelihood function with eleven parameters that can be used to explicitly extract the U-235 content.

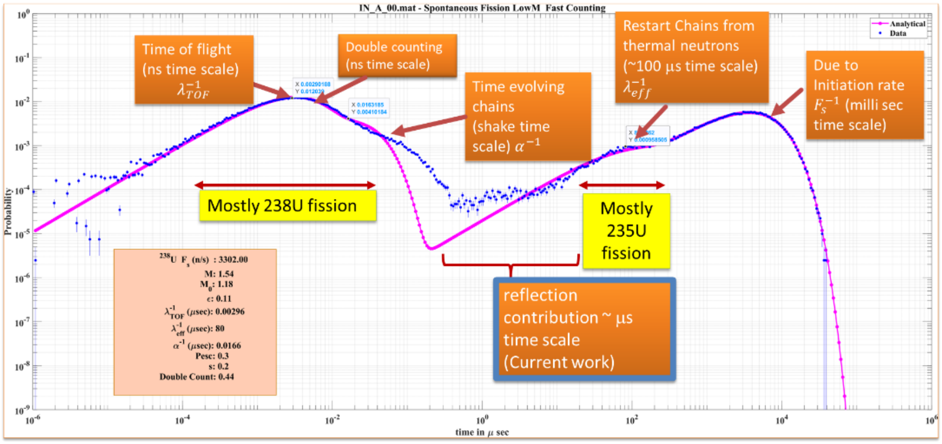

Figure 6. Characteristic profile of the neutron waiting time distribution and the OSANTC likelihood function for function from the eleven-parameter time-evolving fission chain restart theory. Blue dots represent a detected neutron waiting time distribution calculated for a PWR nuclear fuel assembly in the FNCL instrument in Geant4.

The LLNL team has implemented the neutron waiting time distribution analysis and the likelihood function fitting onboard the FNCL computer as a part of the parallel analysis workflow. This enabled processing of experimentally acquired data with an example shown in Figure 7. Here, the neutron waiting time distribution was obtained with the FNCL instrument from a laboratory sample containing ~220 g of U-235. The fit accuracy supports the theoretical assumptions about the system properties and encourages further development. Although the OSANTC solver does not currently account for the effects of neutron poisons present in the system, the underlying theory does account for this complication.

AmLi source replacement with a DD neutron generator

The feasibility of coupling the FNCL instrument with a DD neutron source as a replacement for the AmLi isotopic source during active interrogation measurement was investigated using a portable Thermo-Scientific P385 generator available at LLNL. This DD machine is implemented as a 4 x 30-inch cylindrical head that was coupled with the FNCL instrument using the VVER-1000 front shielding block installed in a reverse orientation as shown in Figure 7. In this configuration, all parasitic X-rays produced near the deuterium target of the neutron generator were effectively shielded. As a result, the FNCL detector cells registered tolerable neutron and gamma-ray count rates and an overall reliable performance during DD source activations in a wide range of neutron outputs. Nuclear material samples were evaluated in the FNCL instrument with the DD generator operating in a continuous mode with a wide range of neutron production rates between approximately 2e5 and 1e7 neutrons per second. In all instances, the FNCL detector arrays and data acquisition system performed reliably, without any indication of an excessive pile-up and saturation. Calibration curves obtained using laboratory material samples with U-235 content between 11 g and 380 g demonstrated monotonic scaling with the fissile material content.

Figure 7. Experimental setup for the Thermo Scientific DD generator coupling with the FNCL instrument using the reverse orientation of the VVER-1000 front shield block.

The FNCL measurements of laboratory samples with the DD neutron generator were processed with the native FNCL software, as well as the GMM-based analysis workflow. Figure 8 illustrates the doubles rate scaling with the U-235 mass in the sample for both analysis approaches. Overall, the experimental measurements confirmed the feasibility of replacing an AmLi source with a DD generator for the FNCL active interrogation. The existing instrument software can handle the resulting event rates on the detectors and appears to maintain efficiency of neutron and gamma-ray discrimination and pile-up rejection. Safe and reliable operations of the DD generator-driven FNCL measurement setup can be confidently implemented for field use by the IAEA.

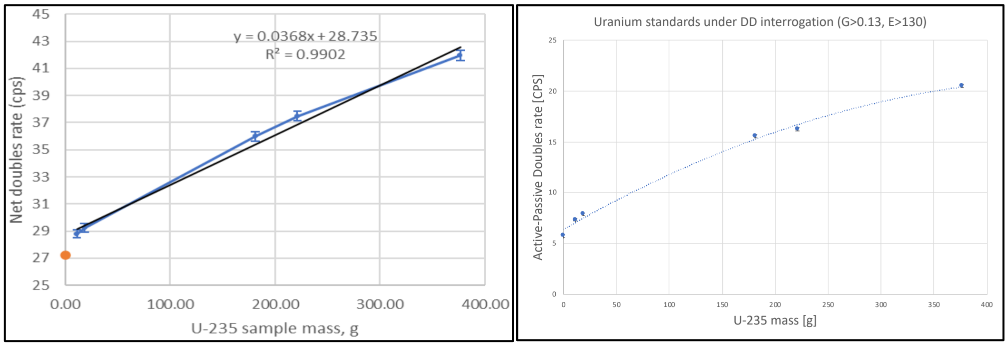

Figure 8. Net neutron doubles rate as a function of U-235 mass in laboratory samples measured with the FNCL instrument coupled with a Thermo Scientific DD neutron generator. Results on the left were obtained with the native software. Data for the plot on the right was reduced using GMM-based analysis software. The generator was operated at 40 uA beam current and 70 kV accelerating voltage. Error bars represent statistical uncertainty only.

Summary

A multi-disciplinary team at LLNL is investigating potential improvements to the current FNCL instrument prototype focusing efforts on optimizing configuration of the detector panels, improved waveform processing methods, self-contained response analysis, and AmLi interrogation source replacement with a DD neutron generator. This work is informed by a detailed instrument response modeling capability implemented in both MCNP and Geant4 transport codes. Laboratory experiments with the FNCL instrument prototype and a set of nuclear material samples are conducted to inform and validate these investigations. At present, the effort has converged on the following findings and accomplishments:

- Commissioned two new FNCL panels that implement a span of candidate hardware design options (scintillation materials, number and size of individual cells, scintillation light readouts). The final determination of the optimal cell configuration will be completed after a series of experimental measurements and cell performance evaluations relative to the baseline FNCL instrument.
- Implemented a parallel waveform processing methodology on the FNCL data acquisition computer based on the Gaussian Mixture Model with improved particle discrimination and pile-up rejection. Based on user input, the algorithm interprets the measured data in a neutron time-of-arrival format, neutron waiting time distribution, or reduces it to the net rate of double coincidence counts.
- Completed initial implementation of the OSANTC likelihood function for the FNCL-produced neutron waiting time distribution as a key approach for self-contained U-235 content determination. Investigated effects from neutron restarts, random neutron contributions, and other system components. The follow-up work will focus on the impacts of neutron poisons.
- Demonstrated the feasibility of replacing an AmLi active interrogation source with a DD generator for the FNCL measurements. The baseline FNCL detector arrays and analysis software demonstrated robust capability to handle elevated event rates at individual detectors as well as maintained efficiency of neutron and gamma-ray discrimination and pile-up rejection.

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