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INITIAL CHARACTERIZATION OF A DD NEUTRON GENERATOR–DRIVEN FAST NEUTRON COINCIDENCE COLLAR

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ABSTRACT

For more than 30 years, the quantitative assay of the ²³⁵U content of light water reactor fresh fuel assemblies relied on measuring coincidence neutrons from fissions induced by an Am(Li) neutron source using ³He based detectors. The Fast Neutron Collar (FNCL), developed by the International Atomic Energy Agency, replaces traditional ³He proportional counters with an array of liquid scintillator detectors arranged about the fuel assembly to provide improved measurement precision and reduced sensitivity to gadolinium poison rods. The FNCL still relies on Am(Li) neutron sources that are no longer commercially available. This work examines the replacement of Am(Li) sources with a commercial offthe-shelf deuterium–deuterium (DD) neutron generator. In addition to mitigating supply concerns, the neutron generator offers advantages in measurement precision and potential automation of sequential passive/active neutron measurements. This paper presents the initial performance results for both the integrated DD/FNCL and Am(Li)/FNCL assays of compact depleted uranium, low-enriched uranium, and highly enriched uranium standards along with an estimate of the expected performance for fresh fuel assemblies.

INTRODUCTION

The Uranium Neutron Coincidence Collar (UNCL) [1] is an active neutron interrogation system used to provide the 235U linear density of fresh fuel assemblies during routine inspection activities in support of international safeguards [2]. The UNCL is rectangular in configuration, three sides of which are arrays of ³He proportional counters embedded in high-density polyethylene. The fourth (active) side contains the Am(Li) neutron source, which is also embedded in high-density polyethylene. During use, the collar is placed around the fuel assembly bringing the Am(Li) neutron source into close proximity with the fuel assembly while encompassing three of the four sides with the neutron detector array. Neutrons emitted from the Am(Li) source induce fission within the fuel assembly, resulting in the emission of additional neutrons. Neutron coincidence counting is employed to distinguish the induced fission from the interrogating source events. The ²³⁵U linear density (i.e., mass ²³⁵U per unit length of fuel assembly) is inferred from the observed neutron coincidence rate, and the result is compared with declared value for the fuel assembly.

The UNCL is based on ³He thermal neutron detectors and provides a modest neutron detection efficiency of about 12% for 252 Cf spontaneous fission neutrons. Because it is based on thermal neutron detection, its characteristic die-away time of 50 µs requires a comparatively long coincidence gate width, typically 64 μ s. The high count rate from the Am(Li) interrogating neutron source coupled with the long coincidence gate leads to high accidentals coincidence rates, which ultimately limit the measurement precision achievable with the UNCL. The UNCL has been used in the field for safeguards for 40 years, and during that time the design and components have changed little. However, sensitivity to poison rods and long assay times required for fast mode measurements have resulted in numerous research efforts to

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provide an improved system. Ongoing UNCL developments fall into three categories: improved ³He designs, alternative detector technologies, and alternative interrogation methodologies.

Detectors with higher efficiency or faster response times could reduce the needed assay time. Higher efficiency variants of the ³He design are possible, and alternative detector technologies such as the boron lined straw detectors and liquid scintillation counters may also offer improved assay performance [3]. Recent work at Los Alamos National Laboratory [4] has investigated the use of a higher efficiency ³He proportional tube based UNCL. The Euratom Fast Collar provides neutron detection efficiency of 15.7% die-away time of 28 µs for a 252Cf point source compared to 12.5% and 50 µs for the UNCL-II. The Euratom Fast Collar is expected to provide improved measurement precision in shorter count times and will have lower sensitivity to the presence of poison rods.

The Am(Li) neutron sources required by the UNCL system have become difficult to obtain commercially. Alternative neutron interrogation sources are also a potential area for development for the UNCL measurement. Some preliminary work has been performed. For example, proof of concept measurements have been performed using a DD neutron generator in place of the Am(Li) source [5]. The use of ²⁵²Cf as an alternative source has been examined by Menlove et al. [6], and Root et al. have begun investigation of a high efficiency passive counting as an alternative to the UNCL for light water reactor fuel [7].

The Fast Neutron Collar (FNCL) [8–10] (Figure 1) has been introduced as alternative for the UNCL measurement of low-enriched uranium fuel assemblies. The FNCL, available from CAEN Industries, replaces the ³He detector assembly with 12 EJ-309 liquid scintillation neutron detectors. Unlike the UNCL, the FNCL is based on fast neutron detection, allowing much shorter coincidence gating, resulting in improved measurement precision by reducing the accidental coincidence rate. Like the UNCL, the FNCL relies on Am(Li) to supply the interrogating neutron flux. However, because the neutrons emitted by the Am(Li) source have an average energy of only a few hundred keV, most of these neutrons fall below the detection energy threshold of the scintillation detectors. That is, the detector is insensitive to the interrogating neutron source, further reducing the accidental coincidence rate and improving the measurement precision. Measurements by Beaumont et al. indicate that the FNCL provides significant improvement in measurement precision, reducing the required measurement times by a factor of 12 relative to the standard UNCL measurement. Their measurements using the FNCL suggest that measurement biases due to the presence of Gd poison rods is also reduced by factor of 3 from 9% to 3%.

Figure 1. Photograph of the CAEN FNCL detector assembly (*left***), the acquisition electronics (***center***), and an isometric view showing the arrangement of the 12 EJ309 neutron detectors [10] (***right***).**

The accidentals coincidence rate associated with the interrogating neutron source and the long coincidence gate width used with ³He based neutron counting systems limits the measurement precision achievable with UNCL. Increases in the interrogating neutron source strength beyond 1E5 n/s offer little to no improvement in measurement precision. The coincidence gate typically applied with the scintillation based neutron detectors used with the FNCL (60 ns) is roughly 1/1,000th the width of that used with the 3 He systems (64 µs), which in principle allows the use of much larger interrogation sources and further improvement in the achievable measurement precision. However, the required Am(Li) sources are no longer commercially available, and sources for new systems are typically salvaged from existing measurement systems. Increasing the interrogating source strength using larger or multiple Am(Li) sources is somewhat problematic, and obtaining the additional improvement in FNCL measurement precision will require an alternative neutron source. Coupling the FNCL with a DD neutron generator will address the source supply limitation as well as provide significantly higher interrogating neutron flux. Commercial off-the-shelf DD neutron generators are available with typical neutron yields up to 2E6 n/s, 40 times greater than available from a single Am(Li) source.

DD Neutron Generator

The DD neutron generator was selected over the more common deuterium–tritium (DT) generator to minimize induced fissions in 238U. Although the 2.5 MeV neutrons from the DD reaction and 14 MeV neutrons from the DT reaction both exceed the fission threshold in ²³⁸U, it is a much simpler task to minimize sensitivity to 238U by the interrogation source if the lower energy DD neutron generator is used. Additionally, the lack of ³H simplifies shipping, and the lower neutron energy requires less additional shielding.

Two DD neutron generators will be considered during this evaluation. The first is a traditional DD neutron generator, the MP320, manufactured by ThermoFisher [11] (Figure 2). The generator produces up to 2E6 n/s in both steady state and pulsed operating modes. The second unit is the nGen350 [12] (Figure 3) currently in development by Starfire Industries for use with the FNCL. The neutron production point of the nGen350 is much nearer to the end of the accelerator tube allowing greater flexibility for integration with the FNCL. The nGen350 operates only in steady state mode with a maximum yield between 1E6 and 2E6 n/s. However, the nGen350 has an external neutron detector assembly to provide active stabilization of the neutron yield. The nGen350 had not yet been received at the time of this writing, so the potential performance of the nGen350/FNCL combination has only been examined by simulation at this time. A comparison of the generator characteristics is provided in Table 1.

Generator Make/Model	ThermoFisher Scientific MP320 [9]	Starfire nGen350 [10]			
Type of generator	DD.	DD			
Maximum emission rate	2×10^6 n/s	\sim 2 × 10 ⁶ n/s			
Neutron energy	2.48 MeV	2.48 MeV			
Output stabilizer	NA.	Active feedback (with external detector)			
Stability		$< 0.1\%$ variation after warmup			
Steady state/pulsed	B oth	Steady state only			
Pulsed mode					
Frequency range	250-20 kHz	N/A			
Duty cycle	$5\% - 100\%$, 5 µs minimum pulse width	N/A			
Generator tube dimensions					
Diameter	12.06 cm	9.0 cm			
Length	55.88 cm	50.0 cm			
Target line	13.97 cm	\sim 1.5 cm			
Weight	11.3 kg	11.45 kg			

Table 1. DD neutron generator characteristics

Figure 2. Photograph of the MP320 controller (*left***) and the detached MP320 DD neutron generator tube (***right***).**

(*right***).**

Benchmark Measurements

No representative or surrogate fuel assemblies were available for performance testing at this stage of the project. Instead, benchmark measurements for the FNCL coupled with both the Am(Li) source and the MP320 generator were performed using a series of low-enriched uranium and HEU items to validate MCNP [13] simulations for the fuel assemblies. An example of the testing arrangement of the FNCL using Am(Li) interrogation sources is illustrated in Figure 4. The uranium oxide standards were placed on a low mass stand at the vertical center of the assay cavity. The testing arrangement of the MP320/FNCL combination is illustrated in Figure 5. The MP320 has been mounted in an high-density polyethylene adapter assembly to position the neutron production target line at the vertical midpoint of the FNCL. The module also affords a degree of personnel shielding and houses a ³He flux monitor tube.

Figure 4. Illustration of the measurement arrangement of the uranium oxide containers in the FNCL using Am(Li) isotopic interrogation sources with the FNCL in the pressurized water reactor (*left***) and boiling water reactor (***right***) configurations.**

Figure 5. Screenshot of the MCNP input file for the MP320/FNCL test configuration for small containers (*left***) and a photograph of the assembled FNCL with the MP320. The cylindrical U3O8 items have been laid on their sides.**

Initial measurements included examination of the passive response of the FNCL to 252Cf, 240Pu, and Am(Li). Table 2 provides the measurement results from several isotopic sources positioned in the center of the assay volume. These measurements were repeated but with the source placed inside a lead pig (wall thickness \sim 1 cm) to examine the gamma-ray sensitivity of the FNCL. From these results it is apparent that the pulse shape discrimination is not adequately rejecting the gamma-ray contribution as currently configured. However, from the doubles rates, we are able to infer that the summed detection efficiency for fission neutrons in the FNCL is 23%. The detection efficiency for Am(Li) neutrons is less than 1% such that the interrogation source introduces only a minimal interference to the coincidence assay.

Note: An oddity of the FNCL analysis software is that count rates are reported as an average over the 12 detector modules rather than as the sum. The values presented in each of the tables in this work represent the summed rates.

The performance of the FNCL in active mode was first examined using two Am(Li) neutron sources with combined yield of approximately 98,000 n/s. The assays consist first of a passive measurement followed

by the active measurement, and each have a duration of 300 s. Table 3 provides a comparison of the measurement and predicted coincidence rates for the New Brunswick Laboratory (NBL) container measurements using Am(Li) sources with the flat active panel shown in Figure 4. Similar measurements using a standard UNCL have been previously performed [5]. Comparison with the earlier measurements confirms the improvement in measurement precision gained by use of the scintillation detectors. For these simple container measurements, equivalent measurement precision was achieved by the FNCL in 1/12th the time required by the UNCL measurement.

	Approx.	Mass	Measured Rates				Simulated			
	Yield (n/s)	$^{235}U(g)$	Singles (cps)		Doubles $(1/s)$		Net Doubles $(1/s)$		Doubles $(1/s)$	
Empty	$1.0E + 0.5$	0.0	161.3	± 2.7	5.3	± 0.7				
Blank Can	$1.0E + 0.5$	0.0	159.2	± 2.8	4.5	± 0.8	-0.8	± 1.0		
Canl	$1.0E + 05$	39.2	184.0	\pm 3.0	9.3	± 0.9	4.0	\pm 1.1	4.8	± 0.1
Can2	$1.0E + 0.5$	102.1	223.7	± 3.2	17.6	± 1.0	12.3	\pm 1.2	10.4	± 0.1
Can3	$1.0E + 0.5$	183.7	256.9	\pm 3.5	22.0	± 1.1	16.7	\pm 1.3	16.1	± 0.1

Table 3. FNCL Active Measurement Results for the Am(Li) interrogating neutron sources.

The measurement performed to characterize the Am(Li)/FNCL system were also performed for the MP320/FNCL combination. The same standards, geometries, and count times were used to simplify the comparison. Examples of the initial active measurement using the MP320 generator are presented in Table 4.

Calculation of the net doubles rate for the development of a calibration curve was complicated by the magnitude and variability of the coincidence background. Assay of a measurement blank was required to provide a proper measurement reference for subtraction of the large active background doubles rates. In addition, it was necessary to normalize the measured rates to account for drifting of the MP320 neutron output and separately subtract the accidental coincidence contribution as it varies from run to run. This analysis is not currently implemented within the FNCL operating software and is performed offline.

With the MP320 generator producing \sim 1.4E6 n/s a coincidence background of 300 cps/detector was observed. From the observed singles rates, the 60 ns coincidence gate would produce an accidental coincidence contribution to the background doubles rate of 10–20 cps/detector. Most of the background coincidence rate is believed to be caused by multiple detections of a given neutron. We are presently evaluating the effectiveness of the FNCL crosstalk filter and methods to reduce the large coincidence background. During the course of this evaluation, we also noted some data acquisition issues with the system; for instance, one or more of the 12 detectors may occasionally drop out leading to unpredictable count rates. An example of such an occurrence is included in Table 4 to illustrate the impact on the measurement.

Expected Performance for Fuel Assemblies

Despite the challenges of the benchmarking measurements and the conclusion that modifications to the neutron generator interface modules will be required, it is still instructive to examine the potential performance of the DD/FNCL for the measurement of fresh fuel assemblies. The simulated assay using the FNCL of a collection of 15×15 and 17×17 light water fuel assemblies were modeled using MCNP [13]. The differences in the neutron interrogation configurations are shown in Figure 6. The Am(Li) source holder and MP320 interface module represent the existing instrumentation, although the nGen350 module is notional and configured to provide minimal moderation to the interrogating flux. The nGen350 interface module will be significantly altered from this simplistic model.

Figure 6. Illustration of the three active interrogation configurations considered for this preliminary analysis using Am(Li) isotopic sources (*left***), the MP320 DD generator (***middle***), and the nGen350 DD generator (***right***).**

It is important not only to understand the expected detection efficiencies and count rates from the assay system, but also to understand the response uniformity and how penetrating the measurement is. Figure 7 presents the fission maps (the fission rate from each fuel pin was determined and plotted as a function of position within the assembly) for a 17×17 pin fuel assembly with ²³⁵U linear density of 65 g/cm. This figure illustrates that placement of the neutron emission point in close proximity to the assembly as with the nonoptimized nGen350 model results in a very localized interrogation of the assembly and that a standoff distance of several cm will be necessary.

Figure 7. Comparison of the total induced fission rates as a function of pin position within a 17 × 17 fuel assembly from an Am(Li) isotopic (*left***), MP320 DD neutron generator (***middle***), and the nGen350 DD neutron generator (***right***).**

From these simulations basic response functions relating the coincidence rate and the linear density of the fuel assemblies can be constructed. As an example, Figure 8 provides a comparison of the simulated response function for the MP320/FNCL system with a measured UNCL fast mode calibration. The slope of the MP320/FNCL response is less steep than the UNCL response because of the increased sensitivity to 238U when using the DD generator. Based on the response curve and simulations for a variety of other poisoned and non-poisoned assemblies, the MP320/FNCL can be expected to perform as well as the Am(Li) based measurement system in terms of measurement precision. The measurement interferences discussed above will have to be addressed to exceed that performance.

Figure 8. Comparison of the MP320/FNCL simulated response as a function of 235U content (neutron yield 1.4E6 n/s) with the measured response of the traditional fast mode Am(Li) UNCL system.

Because the DD generator emits 2.48 MeV neutrons, we expect a greater sensitivity to the ²³⁸U component of the items than when using Am(Li). On a per gram basis, even with the 2.48 MeV interrogating neutrons, the ²³⁵U fission signal is 14 times greater than that of ²³⁸U. However, for the lowenriched fuel assemblies of interest the ²³⁸U mass is 20 times greater than that of the ²³⁵U, and the contribution to the observed doubles rates will be comparable. Various moderator/absorber combinations are under investigation to attempt to tailor the interrogating neutron energy distribution to reduce the sensitivity to ²³⁸U. Table 5 compares the estimated relative fission rates for a variety of items and enrichments for the FNCL and UNCL, illustrating the need to lower the interrogating neutron energy.

Item	Enrichment	Interrogating Source/System				
	(%)	Am(Li)/FNCL	MP320/FNCL	Am(Li)/UNCL		
Can4	2	0.022	0.745			
Can1	20	0.011	0.227			
Can2	52	0.004	0.067			
Can3	93	0.000	0.006			
17×17 array, 20 g ²³⁵ U/cm	1.54	0.089	0.537	0.118		
17×17 array, 60 g ²³⁵ U/cm	4.63	0.082	0.373	0.089		

Table 5. Uranium-238 fraction of total fission event rate.

The distributions of fission events as a function of pin position within a fuel assembly are shown in Figure 9. The Am(Li) interrogation induces relatively few 238U fission events and a relatively uniform interrogation of the fuel assembly. Because there is insufficient moderation of the interrogating neutron flux, the two DD generator configurations produce significantly more ²³⁸U fission events and a less uniform interrogation of the assembly.

Figure 9. Fission maps for a 17 × 17 fuel assembly for the different neutron interrogation sources considered for the FNCL. The top row shows the maps for fission in induced in 235U, and the bottom row shows the maps for fission induced in 238U. Note, the change in scale for the unoptimized nGen350 simulations.

DISCUSSION

The interrogating neutron flux from the DD generators must be tailored to reduce the sensitivity to 238 U while increasing the sensitivity to ²³⁵U. In other words, the average neutron interrogating will be reduced by the addition of moderating/reflecting layers about the generator. This tailoring will also serve to reduce the detection efficiency of the interrogating neutrons improving the measurement precision.

The DD generators produce a significant X-ray exposure rate when operating and may be adversely affecting performance. detectors. Optimization of the gamma-ray shielding has resulted a factor of 2 reduction in total event rates and a 25% decrease in non-pileup events (e.g. 25% of the "neutron signal" were mischaracterized gamma-rays).

An interface module for use with the nGen350 DD generator is being designed to provide a more uniform interrogation as a function of position within the fuel assembly. The interface module will also serve to lower the average interrogating neutron energy to reduce the relative ²³⁸U fission rate.

Stability issues possibly associated with the FNCL data acquisition setup have resulted in large nonstatistical variations in count rates from measurement to measurement. That is the count rates have been observed to shift 20% from one assay to the next. Work to resolve this issue is in progress.

Evaluation of the experimental data and MCNP simulations performed to date indicates that use of the MP320 generator in its current configuration would provide equivalent measurement performance to that provided by the existing Am(Li)/FNCL system. To provide significant performance improvement will require the use an optimized flux tailoring module, improvements to the crosstalk rejection algorithms and generator yield stabilization (provided by the nGen350) to reduce the background doubles rates.

CONCLUSION

Evaluation of the potential use of a DD neutron generator as the interrogating source for the assay of fresh light water reactor fuel assemblies with the FNCL is currently underway. Measurements to date using an MP320 generator have served to identify potential measurement interferences, which will be addressed as the nGen350 neutron generator is adapted for use with the FNCL.

Several measurement interferences have been identified (e.g., gamma sensitivity, stability, crosstalk, and ²³⁸U sensitivity) and efforts to minimize their impact are underway. The majority of the expected improvements will come from simple mechanical changes such as the introduction of additional lead shielding and optimized generator interface module. These will be addressed with the integration of the nGen350 DD generator.

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