Neutron Rodeo Phase II Final Report

Strategic Security Sciences Division

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I. Introduction

The ability to perform nondestructive assay (NDA) of fresh fuel assemblies with high precision in reasonable measurement times is significant for nuclear nonproliferation and safeguards. The current methodology used by the International Atomic Energy Agency (IAEA) for NDA of fresh fuel assemblies involves a He-3 based Uranium Neutron Collar (UNCL) system, which is an active coincidence counting system designed to measure time-correlated neutron doubles from induced fissions within the assembly [1]. The UNCL system provides an easily implementable process of quantifying the ^{235}U linear density of the fresh fuel assemblies; however, operational limitations of the UNCL include long assay times and adverse sensitivity to poisoned assemblies [2]. This has engendered investigation in alternative detector materials for fast-neutron counting to rectify some of these current limitations in the UNCL. The potential improvements of using fast-neutron organic scintillators (e.g. liquid scintillators, stilbene, PSD-capable plastics, etc.) was compared directly to the original Uranium Neutron Collar (UNCL) in-detail during the initial Advanced Neutron Detection Technology Rodeo [3]. The simulation results showed that these alternative detector materials can provide an improvement in the assay time required to reach a suitable precision due to both their lower sensitivity to the interrogation source neutrons (i.e. AmLi neutrons) and their inherently shorter coincidence gate widths. The IAEA has developed a liquid scintillator-based Fast-Neutron Collar (FNCL) [2] as a potential replacement system for the UNCL.

The FNCL is an active coincidence counting system that uses EJ-309 liquid scintillators (LS) for fast-neutron coincidence counting. There are two configurations in the current FNCL, which include a 12-LS system with two AmLi interrogation sources, and an 8-LS system with four AmLi interrogation sources. Each LS cell has a dimension of $10 \times 10 \times 10 \text{ cm}^3$ and has a rectangular geometry. For this report, the 12-LS configuration was used as the benchmark simulation model. Figure 1 shows a diagram of the FNCL system and figure 2 shows an exploded schematic of a single panel. As shown in Figure 1, the FNCL is a three-panel system with a polyethylene bank that holds the two AmLi interrogation sources. Figure 2 shows the components of a detector panel, which is lined with a cadmium and lead liner of 1 mm and 1 cm thickness, respectively. The cadmium liner is used to perform fast-mode NDA on fresh fuel assemblies, where the thermalized neutrons ($< \sim 0.5 \text{ eV}$) outside of the assembly are effectively captured by the liner and are unable to induce subsequent thermal-neutron induced fissions. The lead liner provides suitable gamma-ray shielding to mitigate the high photon flux from the assembly and interrogation sources. This is particularly important as LS and all other organic detector materials are sensitive to neutrons and gamma rays, and a high photon flux degrades the ability to discriminate between the two particle types.

Figure 1. Schematic of FNCL detectors in the three-panel configuration (left) and photograph of the FNCL (right). Four detectors are enclosed in each of the three modular panels.

Figure 2. Exploded schematic of the IAEA FNCL showing the HDPE between detector, the cadmium and lead liners, and the aluminum enclosure

II. Goals and Deliverables

The objective of the Advanced Neutron Detection Technology Rodeo Phase II, hereafter referred to as the Rodeo Phase II, was to investigate any potential improvements in the performance of the current FNCL through fundamentally different interrogation techniques or use of alternative detector materials. The IAEA is currently investigating the potential benefits from the replacement of EJ-309 with other fast-neutron detectors. Simulation case studies were conducted to isolate the benefits from the different scintillator materials. These initial studies provide an idealized direct comparison of the candidate scintillator detector materials.

Each group was then tasked to design a practical and realistic system with various detector types and assay techniques to examine the sensitivity to fissile mass, burnable poisons, interrogation depth in a bundle, and performance of NDA on a variety of different fresh fuel assemblies. All the results from each group were then compared directly with the results from a benchmark simulation of the FNCL.

III. Description of Fresh Fuel Assemblies

Each group was given identical sets of fresh fuel assemblies to be used to characterize the system detector responses. The sets of fresh fuel assemblies included:

1. 17x17 calibration assembly set with 264 fuel pins and 25 guide tubes with ^{235}U linear densities (LD) of 0, 15, 20, 25, 35, 45, 55, 60, and 65 g/cm. These assemblies were used to establish a calibration curve comparing the observed neutron doubles rate to the ^{235}U LDs. Each assembly in this set had an assembly width of 21.4 cm, fuel pin pitch of 1.278 cm, pellet density of 10.41 g/cm^3 , pellet diameter of 0.8255 cm, and a zircalloy cladding with a thickness of 0.057 cm. An image of the pin arrangement of the calibration assembly set is shown in Figure 3. Table 1 summarizes the details for each

Figure 3. 17x17 calibration assembly set with 264 fuel pins and 25 guide tubes. There was a total of nine assemblies in this set with varying ^{235}U enrichment from 0% to 5.01%.

calibration assembly (nine total assemblies).

Table 1. 17x17 calibration assembly set with varying ^{235}U enrichments and monotonically increasing ^{235}U linear densities.

Assembly ID	^{235}U enrichment [%]	^{235}U linear density [g/cm]
Calib 0	0.00	0
Calib 15	1.16	15
Calib 20	1.54	20
Calib 25	1.93	25
Calib 35	2.70	35
Calib 45	3.47	45
Calib 55	4.24	55
Calib 60	4.63	60
Calib 65	5.01	65

- Unpoisoned assembly set that included two 14x14 and two 17x17 fuel assemblies. All four of these assemblies exhibited various fuel assembly parameters including fuel pin pitch, pellet density and diameter, cladding thickness, number and location of guide tubes, and overall enrichment. An image of the assemblies is shown in Figure 4. The details of each assembly in this set are summarized in Table 2 (four total assemblies).

Figure 4. Unpoisoned assemblies. (a) 14x14A, (b) 14x14B, (c) 17x17B.

Table 2. Unpoisoned assembly set with variable fuel assembly parameters

Assembly ID	^{235}U enrichment [%]	^{235}U linear density [g/cm]	No. of fuel pins/guide tubes	Assembly width [cm]	Pellet density [g/cm ³]	Pellet diameter [cm]	Fuel pin pitch [cm]	Zircalloy4 cladding thickness [cm]
14x14A	3.80	37.6	179/17	19.7	10.42	0.875	1.437	0.0617
14x14B	3.13	37.3	176/20	20.6	10.44	0.968	1.499	0.0660
17x17A	3.20	41.3	264/25	21.4	10.52	0.819	1.278	0.0570
17x17B	4.20	54.8	264/25	21.4	10.64	0.819	1.278	0.0570

3. 17x17 partial defect assembly set with various replacement materials. The Low Enriched Uranium (LEU) fuel pins were substituted with depleted uranium (DU, 0.2% enriched), iron, and lead. For each replaced material type, the case where 1, 8, and 40 fuel pins were substituted was simulated. Additionally, the case where fuel pins were substituted uniformly versus those that were substituted from the center of the assembly was also simulated; the centrally-substituted assemblies are denoted as 8a and 40a. All assemblies in this set had a ^{235}U enrichment of 4.00%, an assembly width of 21.4 cm, fuel pin pitch of 1.278 cm, pellet density of 10.41 g/cm^3 , pellet diameter of 0.8255 cm, and a zircalloy cladding with a thickness of 0.057 cm. Figure 5 is an image of the single pin substitution case. Figure 6 and 7 shows the 8 and 8a cases, and the 40 and 40a cases, respectively. Table 3 summarizes the details of the partial defect assemblies (15 total assemblies).

Figure 5. 17x17 partial defect assembly set with a single replaced fuel pin.

Figure 6. 17x17 partial defect assembly set with 8 replaced fuel pins (a) uniformly and (b) centrally distributed.

Figure 7. 17x17 partial defect assembly set with 40 replaced fuel pins (a) uniformly and (b) centrally distributed.

Table 3. 17x17 partial defect assembly set with various replacement materials including Depleted Uranium (0.2% enriched), iron (density = 7.87 g/cm³), and lead (density = 11.35 g/cm³)

Assembly ID	Substituted material	²³⁵ U linear density [g/cm]	No. of fuel pins/guide tubes	No. of substituted pins	Substituted pin arrangement
PD 0	N/A	51.9	264/25	0	N/A
DU 1	Depleted Uranium	51.7	263/25	1	Single Pin
DU 8	Depleted Uranium	50.4	256/25	8	Uniform
DU 8a	Depleted Uranium	50.4	256/25	8	Central
DU 40	Depleted Uranium	44.4	240/25	40	Uniform
DU 40a	Depleted Uranium	44.4	240/25	40	Central
Fe 1	Iron	51.7	263/25	1	Single Pin
Fe 8	Iron	50.3	256/25	8	Uniform
Fe 8a	Iron	50.3	256/25	8	Central
Fe 40	Iron	44.0	240/25	40	Uniform
Fe 40a	Iron	44.0	240/25	40	Central
Pb 1	Lead	51.7	263/25	1	Single Pin
Pb 8	Lead	50.3	256/25	8	Uniform
Pb 8a	Lead	50.3	256/25	8	Central
Pb 40	Lead	44.0	240/25	40	Uniform
Pb 40a	Lead	44.0	240/25	40	Central

4. 17x17 poisoned fuel assembly set containing burnable poisons in the form of Gd-oxide. Assemblies containing 4 and 24 poisoned pins at 6% and 10% wt. concentration of Gd per pin were simulated (four total assemblies). All assemblies in this set had an assembly width of 21.4 cm, fuel pin pitch of 1.278 cm, pellet diameter of 0.8255 cm, and a Zircaloy cladding with a thickness of 0.057 cm. The fuel pins had a ^{235}U enrichment of 4.00 % with a pellet density of 10.41 g/cm³, and the poisoned pins had a ^{235}U enrichment of 2.60 % with a pellet density of 10.21 g/cm³. Figure 8 is an image of the burnable poison assemblies with 4 and 24 poison pins. Table 4 summarizes the details of the burnable poison assembly sets.

Figure 8. 17x17 burnable poison assembly set with (a) 4 and (b) 24 poisoned pins.

Table 4. 17x17 burnable poison assembly set for Gd-oxide concentrations of 6% and 10% by weight.

Assembly ID	Gd concentration [wt%]	^{235}U linear density [g/cm]	No. of fuel pins/guide tubes	No. of burnable poison pins
BP6-4	6%	51.5	260/25	4
BP6-24	6%	49.9	240/25	24
BP10-4	10%	51.5	260/25	4
BP10-24	10%	49.8	240/25	24

In addition to these sets, a ^{252}Cf source (1.33E4 f/s) in the center of the system cavity and the interrogation sources with no assembly inside were also simulated.

As done in the previous Rodeo, each group were to use the calibration assembly set (1) to determine the calibration curve coefficients for fast-mode assay. Once the calibration curve is established, each group was to use their respective curves to estimate the assayed LD of the remaining fuel assembly sets (2-4). The resulting assayed LDs are then compared to the known LD to calculate the mass defect, which quantifies system sensitivity to changes in the fuel assembly of interest. The associated uncertainty in the calculated mass defect is also estimated for a 600 and 1800 second assay,

and included both the contribution from the calibration curve coefficient uncertainty and an assumed systematic uncertainty of 2%.

IV. FNCL with direct replacement of scintillator materials

The first case study aimed to provide a direct comparison (maintaining total system detector volume constant) of different detector materials. To perform a direct comparison of detector materials, the benchmark FNCL simulation geometry was used to directly substitute the EJ-309 LSs with the other alternative detector materials. Figure 9 is the detailed FNCL MCNP benchmark model featuring three panels that house the detectors and a fourth panel that contains two AmLi interrogation sources.

Figure 9. Transverse section of the FNCL model (left). Figure insets show the vertical section (top-right) and the AmLi sources (bottom-right).

a. Simulation Methods

The four different scintillator detector materials that were compared are EJ-309 LS, PSD-capable EJ-276 plastic, trans-stilbene, and PSD-capable small-molecule organic glass. Table 5 summarizes the material properties for each of the compared detector materials.

Table 5. Material properties for each of the candidate scintillator detector materials.

Detector Material	Wavelength of Max. Emission [nm]	Scintillation Efficiency [photons/MeVee]	Hydrogen-to-carbon ratio	Density [g/cm ³]
-------------------	----------------------------------	--	--------------------------	------------------------------

EJ-309 LS	424	12,300	1.25	0.959
EJ-276 plastic	425	8,600	0.92	1.096
Trans-stilbene	390	15,000	0.86	1.160
Organic glass	420	17,400	0.86	1.096

Various light output response models that correspond to each detector material type were used to simulate the response to all sets of assemblies. The neutron light output response function for each detector material was characterized through independent time-of-flight experiments with a ^{252}Cf source. Figure 10 shows the measured neutron light output response for all four detector materials.

Figure 10. Interpolated curves of measured neutron light output responses for each of the candidate scintillation detector materials determined by time-of-flight experiments with a ^{252}Cf source.

The FNCL system was simulated using the MCNPX v2.7e – PoliMi code [1]. MCNPX-PoliMi outputs a detailed interaction log file, i.e. the detector file, which includes all the interactions that occurred in user-selected detector cells.

The detector file includes information, such as the energy deposited inside the detector, the time of interaction, the type of interaction, the energy of the impinging particle, etc., used to reproduce a realistic detector response.

The MCNPX-PoliMi post-processor code, MPPost [2], uses the detector file as input and determines the detector response based on experimentally estimated parameters, such as the quenching function and the energy resolution. Quenching function by Enqvist et al. [3] was used for EJ309 and an exponential fit of the semi empirical quenching function proposed by Monterial et al. [4] was used for stilbene. The quenching functions of EJ276 and organic glass were experimentally derived in the framework of this effort. A detailed description of the method used to determine the light output response to recoil protons is outside the scope of this report.

The energy resolution of a scintillation detector is affected by the material, the detector shape, and light collection devices [5]. In this work, the same resolution function as in Eq. 1 is used for all the materials [3].

$$\frac{\Delta E}{E} = \sqrt{0.113^2 + \frac{0.065^2}{E} + \frac{0.06^2}{E^2}} \quad (1)$$

In Eq. 1, E is the scintillator light output in electron-equivalent units. A list-mode data file of each interaction that results in a detectable pulse was obtained for the simulated assemblies. This time-sorted list-mode data file allows for calculation of the doubles rate and relative uncertainty, and the single rate, as the ratio between the total number of counts and the measurement time.

Gamma-neutron misclassification, pulse-pile up and neutron cross-talk are phenomena that may affect the detector response. Gamma misclassification occurs whenever the pulse shape discrimination method fails to correctly classify a gamma-ray pulse and counts it as a neutron pulse. Cross talk is caused by a single neutron that undergoes multiple subsequent scattering events and induces at least two pulses in multiple detectors, determining a spurious double count. The effects of these phenomena on the final results is discussed in Section III.c.

The performance of different materials were tested for three different types of assemblies, intact assemblies, partial defect assemblies, and burnable poisons assemblies, described in Section III.a and summarized in Table 6.

Table 6. Summary of simulated assemblies, including the ^{235}U linear density range of each assembly.

Type of Assembly	Number of configurations	^{235}U linear density range (g cm^{-1})	Specific features
Intact assemblies	4	37.6-54.8	2 x 14x14, different pin spatial distribution 2 x 17x17, different pin spatial distribution
Partial defect assemblies	16	44 – 51.7	17x17 assemblies with 1, 8, or 40 pins replaced by ^{82}Pb , ^{26}Fe , or DU. Two different spatial distributions are simulated for the 8 and 40 pin-replaced assemblies: (A) uniform replacement and replacement from the center of the assembly.
Burnable poisons assemblies	4	41.8 – 51.5	Assemblies with 4 or 24 pins replaced with ^{64}Gd enriched ones (6% or 10% ^{64}Gd wt%)

Each run was executed at least three times in order to avoid potential bias in the tallied quantities due to an exceeded random number period,

b. Simulation Results

The simulated assemblies aimed at evaluating the performance of the different materials in terms of sensitivity of the estimated fissile mass to the form factor of the assembly (IA), to the partial replacement of fissile material with heavy metals (PDA), and to the presence of gadolinium-enriched rods (BPA).

We used a calibration-based procedure to estimate the ^{235}U linear density of each assayed fuel bundle. First, the system response in terms of neutron doubles was estimated by simulating a series of nine assemblies with increasing ^{235}U linear density, in the $0 - 65 \text{ g cm}^{-1}$ range. A calibration curve resulting from the best fit of the simulated response to these assemblies was then used to estimate the ^{235}U linear density from the neutron doubles count rate, for samples whose fissile content is virtually not known a-priori. This procedure is the same one adopted in the field by IAEA inspectors, as described in the report of the first phase of the Advanced Neutron Detection Technology Rodeo [6]. It should be noted that the ^{235}U linear density of the test assemblies falls within the calibration range; therefore, no extrapolation is needed.

Figure 11 shows the calibration curves for EJ-309, EJ-276, stilbene, and organic glass. As expected, the different materials do exhibit a similar trend, due to the similarity of their quenching functions and ultimately to the same scatter-based neutron detection physics. EJ-309 shows the highest calibration response, despite a lower light output response than organic glass (Fig 11). The lower light output of EJ-309, compared to organic glass, is probably compensated by its higher hydrogen content, i.e. $5.43 \times 10^{22} \text{ atoms cm}^{-3}$ in EJ-309, compared to $4.54 \times 10^{22} \text{ atoms cm}^{-3}$ in organic glass. A variety of functional forms may be used to fit the calibration response. In this work, we used the functional form in Eq. 2, where D is the neutron doubles count rate in s^{-1} and a , b , and c are non-linear least square fitted coefficients.

$$D = a \frac{L}{1+bL} + c \quad (2)$$

Table 7 reports a , b , and c coefficients of Eq. 2 for the simulated materials. A metric of the assay performance of a collar is the mass defect, which is the relative difference of the estimated ^{235}U LD compared to the declared one. This metric is typically reported as a percentage.

Figure 11. Simulated calibration curves of the FNCL featuring EJ309, EJ276, stilbene, and organic glass.

Table 7. Summary of simulated assemblies, including the ^{235}U linear density range of each assembly

Coefficient	EJ-309	EJ-276	Stilbene	Organic glass
a ($\text{cm g}^{-1}\text{s}^{-1}$)	5.20 ± 0.049	3.862 ± 0.02	4.191 ± 0.02	4.394 ± 0.04
b (cm g^{-1})			$1.50 \pm 6.4\text{E-}4$	$1.159 \pm 3.2\text{E-}4$ $1.258 \pm 3.1\text{E-}4$ $1.320 \pm 7.6\text{E-}4$
c (s^{-1})			0.007 ± 0.55	0.006 ± 0.21 0.006 ± 0.22 0.008 ± 0.45

Figure 12 shows the mass defect of the unpoisoned, intact fuel assemblies. An exact assay would result in a mass defect of zero. An accurate estimate is obtained in the case of the 17x17 assembly. Regardless of the detection material, the FNCL is able to accurately estimate the ^{235}U LD of both 17x17A and 17x17B, which feature a ^{235}U enrichment of 3.2 wt. % and 4.2 wt. %, respectively. Conversely, using a calibration curve based on the assay of a 17x17 assembly results in an inaccurate ^{235}U LD estimate of smaller 14x14 assemblies. The LD is overestimated when channels left empty to allow insertion of the control rods are uniformly distributed within the assembly, while it is underestimated in the case when the empty channels are localized in five positions, in groups of four (Section III.a). Overall, the mass

defects estimated by the FNCL models featuring different materials are consistent with each other within $\pm 1 \sigma$ error bar, which includes the combined systematic and statistic (1SD) uncertainties, as detailed in section IV.a.

Figure 12. Simulated mass defect for the intact assemblies.

Figures 13-15 show the mass defect of assemblies with one, eight, or 40 pins replaced with high atomic-mass materials, i.e. lead, iron, and depleted uranium. The mass defect of the PDAs shows the sensitivity of the FNCL to diversion scenarios. The FNCL is always able to detect a diversion of 40 pins, i.e. approximately 14% of the total fissile mass of the assembly, while an assembly with one missing pin cannot be distinguished from a full assembly, in practice. A diversion of eight pins can be detected if the replacement material is either depleted uranium or iron, while the mass defect is within ± 1 SD of a zero mass defect when eight fuel pins are replaced by lead. The higher mass defect in the case of lead is probably due to the lower neutron capture cross section of lead, compared to DU and iron.

Figure 13. Simulated mass defect for the partial defect assemblies, with 1, 8, and 40 fuel rods replaced by depleted uranium.

Figure 14. Simulated mass defect for the partial defect assemblies, with 1, 8, and 40 fuel rods replaced by iron rods.

Figure 15. Simulated mass defect for the partial defect assemblies, with 1, 8, and 40 fuel rods replaced by lead rods.

Partial defect assemblies with a replacement of eight or forty pins were simulated in two different configurations, the first where the pins were removed from the middle of the assembly and the second, A, with uniform pin removal. The relative difference between the mass defect in case of uniform removal, compared to a removal in the center of the assembly, is shown in Fig. 16 and is an estimate of the penetrability of the assay. A negative relative difference confirms that the FNCL is more sensitive to pin removal from the middle of the assembly, rather than a distributed one.

Figure 16. Ratio between mass defects simulated with uniform pin removal and removal from the center of the assembly.

Gadolinium enrichment of the fuel rods unavoidably affects the fission dynamics during active interrogation of the fuel assembly. An average drop in the neutron doubles signal can be observed as the number of Gd-enriched pins increases from four to 24, and the Gd content increases from 6 wt% to 10 wt%, as shown in Fig. 17. With respect to unpoisoned fuel assemblies, an average drop of the neutron doubles signal of 2.2% and 8.4% results from the insertion of 6 wt% and 10 wt% Gd loaded pins, respectively.

Figure 17. Neutron doubles rate as a function of the number of gadolinium enriched rods (6 wt% and 10 wt%) for the FNCL based on (a) stilbene , (b) EJ309, and (c) glass.

Figure 18 shows the mass defect analysis of the four gadolinium-poisoned simulated assemblies and confirms a drop-in count rate. The FNCL is insensitive to a loading of 4 pins at 6 wt% for all the detection materials, except EJ-309.

Figure 18. Simulated mass defect for the burnable poisoned assemblies.

c. Uncertainty analysis

The uncertainty associated with any mass estimate is given by the combination of three different contributions: the first due to systematic uncertainty, the second to counting statistics fluctuations, and the third to the calibration. When performing a calibration-based assay, the ^{235}U linear density is calculated as in Eq. 3, obtained by rearranging Eq. 2.

$$L = \frac{D-c}{a+b(c-D)} \quad (3)$$

$$\sigma_F = \sqrt{\left(\frac{\partial L}{\partial D}\right)^2 \sigma_D^2 + \left(\frac{\partial L}{\partial a}\right)^2 \sigma_a^2 + \left(\frac{\partial L}{\partial b}\right)^2 \sigma_b^2 + \left(\frac{\partial L}{\partial c}\right)^2 \sigma_c^2 + 2\left(\frac{\partial L}{\partial a}\right)\frac{\partial L}{\partial c}\sigma_{ac} + 2\left(\frac{\partial L}{\partial b}\right)\frac{\partial L}{\partial a}\sigma_{ab} + 2\left(\frac{\partial L}{\partial b}\right)\frac{\partial L}{\partial c}\sigma_{bc}} \quad (4)$$

The systematic relative uncertainty, ϵ_S , is estimated as 2% of the gross neutron doubles count rate. This is an experimentally estimated value and accounts for all sources of systematic error, e.g., temperature fluctuations that affect the detectors' response. The relative uncertainty due to counting statistics, $\epsilon_M = \sigma_D/D$, is calculated as the standard deviation of the moments of neutron multiplicity distributions, using the method proposed by Prasad M. et al. [7]. Using a calibration curve to estimate the ^{235}U LD from the simulated neutron doubles rate requires propagating the uncertainty associated

Figure 19. Uncertainties associated with the estimated ^{235}U linear density by (a) EJ309-, (b) EJ276-, (c) stilbene-, and (d) organic glass- based FNCL.

with the fitting coefficients when estimating the ^{235}U LD. This procedure allows to estimate $\epsilon_F = \sigma_F/D$, i.e., the sample standard deviation of the estimated ^{235}U LD due to the appropriateness of the fitting model in capturing all the information in the calibration data. Eq. 4 shows the standard deviation related to the calibration procedure. The detail of the terms in Eq.4 is reported in Appendix A. The total relative uncertainty associated to a measurement, s_{TOT} , is then obtained as the quadratic squared sum of the three terms. Fig 19 shows s_{TOT} , ϵ_s , ϵ_M , and ϵ_F , while ϵ_s is always assumed to be 2% of the neutron doubles count rate.

d. Remarks about gamma-neutron discrimination, pulse pile up and cross talk

Organic scintillators are sensitive to both neutrons and gamma rays. The different decay constants of light pulses produced by neutrons and photons make it possible to discriminate between them by implementing methods based on pulse-shape discrimination (PSD). Compton scattering and neutron scattering on hydrogen nuclei are the two main interactions that allow the detection of gamma-rays and neutrons, respectively. Recoil protons and electrons are the charged particles depositing energy in the scintillator after neutron and gamma-ray interactions, respectively. Light produced by recoil proton excitations of the scintillators is heavily quenched [8]. This phenomenon makes pulse shape discrimination particularly challenging at relatively low neutron energies (< 500 keV) and a material with a good PSD capability should be selected to minimize the misclassification of gamma-ray pulses as neutron pulses, and vice versa [9].

In this work, PSD was performed by calculating the ratio between the digital charge integral of the pulse tail to that of the full pulse. The bounds of the integrals were defined by three parameters in time: the start of the integrals, the tail-stop, and the total-stop. Each parameter was separately optimized for the different materials. The figure of merit (FOM) is a parameter used to quantify the PSD capability of a detector. FOM is defined as in Eq. 5, where δ is the difference between the means of the neutron and gamma distributions in a tail-total ratio histogram (Fig 20), and FWHM_n and FWHM_γ are their respective full widths at half maximum.

$$\text{FOM} = \frac{\delta}{\text{FWHM}_n + \text{FWHM}_\gamma} \quad (5)$$

Figure 21 compares the measured FOM as a function of light output for the materials investigated in this work, for cylindrical detector geometries. Pulse misclassification can be mitigated by operating the detector at a relatively high detection threshold of approximately 650 keV neutron energy deposited by recoil protons. At the selected a detection threshold, the misclassification rate of gamma-ray pulses is approximately 5×10^{-5} per detection.

Figure 20. Scatter density plot of a ^{252}Cf source measured by EJ276 (left) and 100-150 keVee slice of the scatter-density plot (right).

Figure 21. Figure of merit as a function of light output for EJ309, EJ276, stilbene, and organic glass.

Although organic scintillators exhibit a fast response of tens of nanoseconds, pile up of pulses can occur, in the presence of a high fluence rate. Piled-up pulses are prone to be wrongly classified as neutron pulses. Disregarding piled-up pulses has the effect of reducing the actual efficiency of the detector and increasing the assay time to achieve a given uncertainty. In the case of fresh fuel, thanks to the 1-cm thick lead shield between the fuel and the detectors, a count rate of approximately 10^5 gammas s^{-1} is expected, including both gamma-ray and neutron pulses. At this count rate, piled-up

pulses would be approximately 3% of the total number of pulses [5] and therefore negligibly contribute to the total count rate.

Fast organic scintillators rely on scattering reactions to detect neutrons. A neutron can undergo multiple scattering reactions in different detectors and therefore deposit energy above the acquisition threshold in more than one detector. This phenomenon is typically referred to as cross talk and may cause spurious neutron coincidence counts, adversely increasing the total estimated fissile mass. In the simulated FNCL configuration, cross talk coincidences account for approximately 30% of the total number of coincidences. In this section of the work, we aimed at comparing different materials when used in the FNCL and did not discriminate and filter neutron coincidences due to cross talk. In fact, the neutron doubles rate due to cross talk is expected to be comparable for the different scintillating materials, because of their similar composition. Other factors affecting cross talk, such as the neutron energy spectrum and the geometric configuration of the detection system are comparable for the different simulated versions of the FNCL.

V. NDA of fresh fuel assemblies

This study was a collaborative effort aimed to investigate each group's system performance and directly compare the results to the benchmark FNCL simulations. A set of ground rules was established for the design of each group's system in order to maintain some foundation for fair comparison. Each group was to

1. Roughly maintain the form-factor of the FNCL system
2. Use MCNP for the simulation of system response in fast-mode only (i.e. with Cd liners)
3. Use identical fresh fuel assembly inputs
4. Use identical AmLi interrogation neutron source definition
5. Provide validation of simulated detector response with experimental results

Rodeo Phase II featured two different assay techniques for NDA of the fresh fuel assemblies. The first technique is an active assay using AmLi interrogation sources to induce fissions within the assembly and is the current technique implemented in the IAEA FNCL. The typical procedure for active assay using AmLi sources involves three separate measurements. First, the ambient background count rate is measured (no fresh fuel assembly) and is subtracted from any subsequent measurements. Then, the fuel assembly is placed within the cavity and a passive assay (i.e., no AmLi sources) is implemented to measure the doubles count rates from the spontaneous fission of ^{238}U . Lastly, the AmLi sources are introduced to interrogate the fresh fuel assembly and measure the doubles count rates from the induced fission of ^{235}U in addition to the count rates from the spontaneous fission of ^{238}U . The subtraction of the ambient background and passive assay count rates on those measured from the active assay gives a net doubles count rate that is proportional to ^{235}U LD, assuming that the majority of the induced fission occurs on ^{235}U . The AmLi interrogation neutron source energy spectrum was simulated using the Obninsk spectrum provided by the collaboration. Figure 22 shows the simulated energy spectrum from the AmLi interrogation source.

Figure 22. Neutron energy spectrum for the AmLi interrogation source used for all active assay simulations.

The second technique is a passive assay that does not require any interrogation sources. The implementation of the passive assay is predicated on the ^{238}U spontaneous fission neutrons acting as the initiating neutron source for inducing fissions on ^{235}U . Therefore, both the measured singles and doubles count rate would be proportional to the ^{235}U LD. This “self-interrogation” source from the ^{238}U spontaneous fission neutrons would be well-distributed throughout the assembly allowing for improved interrogation penetrability. Furthermore, the absence of a perpetually-emitting interrogation source provides benefits in operational and procedural costs.

a. Detailed System Descriptions

In total, there were five systems that were simulated and directly compared to the benchmark FNCL. Table 8 is a summary of the detector material, interrogation source, and assay technique implemented by each group.

Table 8. Summary of all simulated systems from each group including detector material, assay technique, and number of interrogation sources

Group	Detector Material	Detector Dimensions	Detection Mechanism	Assay Technique
		Number of cells/slabs		
IAEA (Benchmark FNCL)	EJ-309 liquid organic	10 x 10 x 10 cm	Elastic scatter on recoil nuclei	Active assay with 2 X AmLi sources
		12 cubic cells		
LLNL	Trans-stilbene	10.16 x 5.08 cm	Elastic scatter on recoil nuclei	Active assay with 1 X AmLi sources

30 cylindrical cells				
UM	Trans-stilbene	5.08 x 5.08 cm	Elastic scatter on recoil nuclei	Active assay with 2 X AmLi sources
30 cylindrical cells				
SNL/INL	Small-molecule organic glass	10 x 10 x 10 cm	Elastic scatter on recoil nuclei	Active assay with 2 X AmLi sources
12 cubic cells				
LANL1	He-3 tubes embedded in polyethylene	10 x 24 x 54 cm Single detector slab including polyethylene	Capture reaction on He-3	Passive assay with self-interrogation technique
4 slabs				
LANL2	Boron-10 cells embedded in polyethylene	15 x 24 x 54 cm Single detector slab including polyethylene	Capture reaction on Boron-10	Passive assay with self-interrogation technique
6 slabs				

b. UM1: FNCL

UM simulated the EJ-309 based FNCL, in a configuration which resembles as closely as possible the one currently being tested in the field by the IAEA. The first version of the model was provided by the IAEA. The simulated version of the FNCL is based on 12 EJ-309 scintillation detectors. Each detector has the shape of a 10-cm side cell and coupled to 7.6 cm diameter photomultiplier tubes (PMT, ETL type 9821 FLB). The FNCL encompasses three detector panels, each including four detectors. The system can be assembled in two-panel and three-panel configurations. Only the three-panel configuration was simulated in this work. Adjacent detectors are spaced from each other by 1 cm of high-density polyethylene (HDPE) to minimize neutron cross-talk between detectors. The detectors are shielded from the intense gamma-ray flux of fresh fuel by 1-cm of lead. Additionally, a 1-mm Cd liner prevents the diffusion of low-energy neutrons from the assembly and shields the assembly itself by back-scattered thermal neutrons. The cadmium liner surrounds the four sides of the fuel assembly. An external aluminum enclosure was also modeled, which guarantees durability and sturdiness to the assembly. The interrogating AmLi sources are placed inside HDPE slab placed on the side of the fuel assembly free from the detector panels. Two AmLi time-dependent sources were simulated, each having a source strength of 5×10^4 neutrons s^{-1} . Figure 23 shows a visual rendering of the simulated model.

Figure 23. (a) Three-dimensional rendering of the MCNPX-PoliMi model of the FNCL system, including a fuel bundle and (b) its transverse plane.

The simulation was performed using MCNPX-PoliMi, in combination with the post processor MPPost, which converts the energy deposited inside the detector in light pulses and it is thus capable of accurately reproducing the detectors' response. In this case, a list-mode file including all the detected pulses was generated by MPPost and further used to calculate the singles and neutron doubles distribution. The IAEA-FNCL includes sophisticated software that performs post-processing algorithms such as pulse shape discrimination, pulse pile-up rejection, anti-cross talk filter. None of these was implemented in the simulated model, to keep the post processing procedure as simple as possible and consistent with the simulations performed by the other groups.

c. LLNL: Active neutron collar with stilbene and one AmLi source

LLNL simulated a stilbene-based active neutron collar consisting of 30 10.16 cm by 5.08 cm depth cylindrical stilbene cells. Stilbene is an organic scintillator that is known to have excellent pulse-shape discrimination (PSD) capability and high neutron detection efficiency. As the case with all scintillator detectors, the response time is much faster than capture-based neutron detectors (timescale on the order of nanoseconds versus microseconds), which allow for a lower rate of accidental correlated counts thus better measurement precision and lower assay times. Stilbene detectors are nonflammable solid-state crystals making it an ideal candidate for practical use.

Each stilbene detector is coupled to a photomultiplier tube of the same diameter and is housed in a thin aluminum case. Figure 24 is an image of the bare stilbene crystal along with the fully-enclosed detector assembly.

Figure 24. Photograph of 4" diameter by 2" length stilbene crystal (left) and photomultiplier for scintillation light readout (right).

The LLNL stilbene-based active neutron collar has three panels consisting of 10 stilbene detector assemblies each and one panel for the single AmLi interrogation source embedded in polyethylene. The detectors in the panels are held by a rigid low-density foam frame; figure 25 shows a detector panel along with the MCNP simulation model of the full system.

Figure 25. Single panel of the LLNL prototype stilbene neutron collar (left) with 10 stilbene cells. MCNP model of the stilbene collar with a fuel assembly (shown without the poly bank in which it is embedded).

The full-system dimensions are 90 x 63 x 50 cm. The total detector volume within the system is 12,356 cm³ with approximately 2432.2 cm² of detector surface facing the fuel. A Cd liner of 0.5 mm thickness is placed along the walls of the inner cavity. A lead shield of 1 cm thickness is also placed in front of the detectors to reduce the high-photon flux.

The electronics required for power supply and data acquisition can be contained in a single VME crate. The output from each detector is digitized through two 16-channel Sys Struck 3316 digitizers, which can have up to a 16-bit vertical resolution and a 250 MS/s sampling rate. All detectors would be powered by

two 16-channel ISEG high voltage power supply. The data is output as list-mode data that includes the detection time stamp, total pulse integral, pulse shape parameters for PSD, and detector ID number.

The estimated cost and weight of this system including all required electronics and ancillary components is approximately \$325,000 and 150 kg, respectively. The system is cart-portable, and can be assembled within an hour. A 10-stilbene detector panel has been assembled, calibrated, and tested with AmLi, AmBe, ^{252}Cf , D-D, and D-T neutron test sources. Modeling and simulations have been completed for the test sources and the fuel assemblies. Multiple scattering corrections and quench functions have been completed.

d. UM2: Active neutron collar with stilbene and two AmLi sources

The University of Michigan investigated a stilbene-based active neutron collar consisting of 30 5.08 cm diameter by 5.08 cm depth cylindrical stilbene cells. The benefits of using stilbene are the same to those mentioned in the LLNL system description. Mainly, stilbene detectors have excellent PSD capability, they are solid-state, and they have shown good intrinsic detection efficiency for fast neutrons.

Each stilbene cell is wrapped in polytetrafluoroethylene (PTFE) tape and contained in a thin aluminum casing that includes an optical window. Each cell is coupled to an ET 9214b photomultiplier tube (PMT) and is housed in a light-tight detector case; figure 26 shows the full stilbene detector assembly and the detailed MCNP model.

Figure 26. Stilbene detector with photomultiplier inside a 3D-printed case and its MCNP rendering (right).

The system features two AmLi interrogation sources embedded in polyethylene placed in opposite panels. A Cd liner of 0.5 mm thickness is placed along the walls of the inner cavity. A lead shield of 1 cm thickness is also placed in front of the detectors to mitigate the high-photon flux. The full-system dimension is 76 x 40 x 40 cm. The total detector volume within the system is 3,089 cm³ with approximately 608.1 cm² of detector surface facing the fuel. A figure of the simulated system used for this research is shown in figure 27.

Figure 27. The geometry of the simulated model of the UM stilbene collar consisting of 24 stilbene detectors.

Signals from each detector are readout digitized through two 16-channel CAEN V1730 waveform digitizer with a 2-V dynamic range, 14-bit vertical resolution, and a 500 MS/s sampling rate. All detectors are powered by five 6-channel CAEN V6533 HV power supply. On-board data processing includes gamma-ray rejection and digital constant-fraction timing algorithm. The data acquisition software is able to acquire both the full waveform data as well as list-mode data with a maximum data throughput of approximately 110 Mb/s without any losses. The list-mode data gives the detection time, and pulse-integral values used for PSD. The waveform-mode acquisition gives the digitized waveform for each signal in addition to the information provided by the list-mode data. The estimated cost and weight of this system including all required electronics and ancillary components is approximately \$150,000 and 100 kg. The system is cart-portable and can be assembled within one hour.

A 24-stilbene detector system was used to measure the neutron singles and doubles count rate, which was then used to compare to the results from a detailed MCNP simulation. Figure 28 is an image of the 24-stilbene detector system along with the detailed MCNP simulation model.

Figure 28. (a) Photograph of the UM FNMC, including 24 stilbene detectors. (b) Side view of the simulated UM collar. (c) Top view of the simulated UM collar

The comparison of the neutron singles and doubles rate is summarized in Table 9 for three different detection thresholds.

Table 9. Simulated neutron count rates at 50, 75, and 100 keVee threshold.

Detection Threshold	Simulated singles [cps]	Measured singles [cps]	Difference in singles	Simulated doubles [cps]	Measured doubles [cps]	Difference in doubles
50 keVee	2883.17 (0.86)	2857.17 (0.56)	0.91 % (0.04)	306.67 (0.28)	244.95 (0.25)	25.20 % (0.16)
75 keVee	2373.75 (0.78)	2511.14 (0.53)	-5.47 % (0.04)	201.96 (0.23)	186.84 (0.22)	8.09 % (0.17)
100 keVee	2019.34 (0.72)	2145.74 (0.49)	-5.89 % (0.04)	143.41 (0.19)	134.90 (0.18)	6.31 % (0.19)

From the results shown in Table 9, the simulated neutron singles and doubles rates were within 6% and 9% of the measured neutron singles and doubles rates, respectively, for detection thresholds of 75 and 100 keVee.

e. SNL/INL: Active neutron collar with organic glass scintillators and two AmLi sources

Sandia National Laboratories has recently developed low-cost, potentially large-scale, high-performance fast-neutron PSD-capable organic glass scintillators. Compared to other liquid (i.e. EJ-309) or solid (i.e. trans-stilbene) scintillators, an advantage is that organic glass scintillators can be melt cast, which greatly simplifies their manufacture and may a) have lower production costs and b) allow for the fabrication of large detector cells. Equally important to the physical/manufacturing advantages of organic glass is its enhanced neutron detection performance. Notably, in small 2.5 x 2.5 cm right-cylinder geometry, it has both better pulse-shape discrimination capability and higher light yield than trans-stilbene. Initially developed detectors with larger dimensions (5 x 5 cm) tested by Idaho National Laboratory and University of Michigan have continued to show higher light yield with slightly worse PSD capability but an increase in neutron detection efficiency of 15-20%.

The primary potential advantages of the PSD-glass detectors over stilbene or EJ-309 are higher neutron detection, efficiency, ruggedness and reduced cost. Compared to EJ-309, being a solid the PSD-glass is much more robust. The primary current disadvantage of the PSD-glass is that its manufacture is currently implemented at a lab scale for small detector sizes. Development of larger detectors and commercial manufacturing will be needed for the detectors to be feasible replacements for currently available EJ-309 or stilbene detectors. SNL is actively engaged in technology transfer to a commercial vendor, and development of larger size detector samples. For the measurement of fresh fuel, the total PSD-glass detector volume would be 12,000 cm³, the same as the EJ-309 in the FNCL. This volume is composed of twelve 10 x 10 x 10 cm detectors. Four detectors are arranged into a panel on each of three sides of the fuel assembly, with each panel having 400 cm² of detector surface facing the fuel. A Cd liner of 1 mm thickness was used for the fast-mode assay. A lead shield of 1 cm thickness was also placed between the detector and the fresh fuel assembly. Similar to the current FNCL, the system features two AmLi interrogation sources embedded in polyethylene as shown in figure 27. The output from the detectors can be collected by the FNCL CAEN SD7750 in full waveform-mode, or in list-mode using CAEN, Struck or another digitizer.

The portability of the system is the same as the current FNCL. It is expected that utilizing PMTs and organic glass will result in a much more rugged system than with the current EJ-309 LS-based FNCL design. Use of SiPM would significantly improve the robustness of the system.

Experimental time-of-flight (TOF) measurements were performed with 5x5cm right cylinder PSD-glass samples at both the INL and UM. These measurements were performed with ETL 9214B PMTs in both cases, but with different data acquisition systems, ²⁵²Cf sources, energy cutoff levels, and source-to-detector distances. Detection efficiency for ²⁵²Cf neutrons incident on the detectors was determined to be 35% +/-2% in both experiments. MCNPX-PoliMi simulations of the same geometries, using 35keVee energy cut result in a 40% neutron detection efficiency. The peak emission from the PSD-glass is at a

higher wavelength than optimal for the ETL 9214B PMT, resulting in a 7-10% decrease in quantum detection efficiency. This is expected to be the primary cause of the discrepancy, combined with a lower energy cutoff used in the MCNPX-PoliMi simulations. The larger ETL 9821 PMT used in the FNCL has a similar light response as the ETL 9214B, so a 12.5% reduction in neutron response was applied in post processing to the MCNPX-PoliMi results for the PSD-glass FNCL. This reduction in neutron detection efficiency results in a 20-25% decrease in expected detected coincidence rates from that predicted by MCNPX-PoliMi alone.

f. LANL1 & 2: Passive neutron collars with capture-based detectors

Los Alamos National Laboratory simulated two passive neutron collar systems that use ^3He and ^{10}B capture-based detectors. The four major advantages for using a passive neutron collar approach is that a) it does not require AmLi sources that are in short supply, b) simpler and more user-friendly where the present IAEA training protocol is directly applicable, c) smaller and more robust than the scintillator-based systems, and d) in-plant measurement does not require AmLi source handling, transfer, and licensing. In addition to these four practical advantages, the passive neutron assay approach provides calibration curves that are more linear than the active assay approach due to the harder neutron interrogation spectrum. This near-linear calibration curve introduces less error in the estimated ^{235}U LD. Furthermore, both the singles and doubles count rates can be used due to the absence of the active interrogation source.

The ^3He detector system consists of 28 ^3He tubes embedded in high-density polyethylene, designed to fit the form factor of the original UNCL. The benefits of using ^3He tubes for NDA include the high intrinsic neutron efficiency, the insensitivity to gamma-rays, and the practical robustness of the ^3He tubes; the robustness and reliability of ^3He detectors has been thoroughly tested with the previously-used UNCL with decades of trouble-free use under field conditions. Since the assay technique used by the LANL group is passive self-interrogation, the system design does not have a designated panel for the interrogation sources. This panel is instead replaced by another slab of detectors. Figure 29 shows the MCNP model for the passive ^3He detector system along with an image of a single ^3He detector slab.

Figure 29. Visual rendering of the simulated ^3He based collar in transverse (left) and side (middle) sections and a photograph of the assembled system (right).

The full dimensions of the ^3He detector system are 44 x 44 x 54 cm. The total detector volume (including the high-density polyethylene moderator) of the system is 73,440 cm³ with a total detector surface area of 5184 cm² facing the fuel. The system features a Cd liner of 0.5 mm thickness within the walls of the inner cavity. The estimated weight is about 73 kg including all required electronics and ancillary components. The required electronics is identical to the UNCL system and includes 4 PDT-10A amplifiers for each panel of detectors, and the JSR-15 for data collection and power supply. The acquired data is in the form of list-mode data giving a time stamp for all detected neutrons with a maximum data count rate of 1 MHz without dead time losses. Similar to the ^3He detector system, LANL also investigated ^{10}B detector system. The ^{10}B detector system consists of six detector slabs each containing a total of 40 sealed cells coated with ^{10}B . Thin high-density polyethylene (8 mm thick) moderator between each cell is placed to replenish the thermal-neutron flux. Figure 30 shows the MCNP model for the passive ^{10}B detector system along with an image of a single ^{10}B detector slab.

Figure 30. Visual rendering of the simulated B-10 based collar in a top (left) and side (middle) views and a photography of the assembled system (right).

The full dimensions of the ^{10}B detector system are 56 x 46 x 54 cm. The total detector volume (including the high-density polyethylene moderator) of the system is 129,000 cm³ with a total detector surface area of 5184 cm² facing the fuel. The system also features a Cd liner of 0.5 mm thickness within the walls of the inner cavity. The estimated weight is about 131 kg including all required electronics and ancillary components. The required electronics is similar to the ^3He detector system but includes a PDT preamplifier on each cell. The acquired data is output as list-mode similar to the ^3He detector system with a maximum data count rate of 10 MHz.

The individual detector slabs were benchmarked using a ^{252}Cf source. This benchmark establishes the threshold for the ^{10}B detector slabs. The ^3He MCNP simulations were determined based on prior collar

Figure 31 Single and double count rates as a function of ^{235}U linear density for the LANL passive systems based on He-3 and B-10.

systems that were previously benchmarked. Figure 31 shows the benchmarked MCNP calculations of the calibration assembly set for the ^3He and ^{10}B detector systems.

VI. Results: comparison of all simulated systems

In this section, we report and compare the results obtained simulating the response of six different collar system designs in assaying 33 types of fuel assemblies; the six collar system designs are summarized in Table 10. Compared to the first phase of the Neutron Rodeo, all the systems are operated in so-called fast mode, i.e. a 1-mm Cd liner is placed between the fuel and detectors. This addition allows the filtering of the thermal neutrons (<0.5 eV) that would be scattered back from the detectors to the fuel assembly. Fast-mode operation minimizes the thermal neutron portion of the spectrum inside the assembly. This feature is particularly important when assaying the LD of assemblies containing burnable poisons, i.e., materials with a high capture cross-section for thermal neutrons, as it will be detailed in Section VI.d.

The detection systems, described in section V, fall in two main categories: active systems, which rely on external AmLi sources to induce fission reactions in the assemblies, and passive systems, which exploit neutrons emitted by ^{238}U spontaneous fissions to induce fission reactions on ^{235}U .

Table 10. Summary of simulated systems.

Group	Data Acquisition Electronics	Data Structure	Gamma-ray Shielding	Cd liner thickness	Portability
IAEA (Benchmark FNCL)		List-mode data with time stamp, pulse-shape parameters, and channel number Waveform-mode for acquisition of full digitized waveforms	1 cm lead	1 mm	Cart portable
LLNL	2 x 16ch Sys Struck 3315 digitizers	List-mode data with time stamp, pulse-shape parameters, and channel number	1 cm lead	0.5 mm	Cart portable
UM	2 x 16ch V1730 CAEN digitizers	List-mode data with time stamp, pulse-shape parameters, and channel number Waveform-mode for acquisition of full digitized waveforms	1 cm lead	0.5 mm	Cart portable
SNL/INL			1 cm lead	1 mm	Cart portable
LANL1	JSR-15 multiplicity shift register	List-mode data with neutron time stamp	None	0.5 mm	Cart portable
LANL2	JSR-15 multiplicity shift register	List-mode data with neutron time stamp	None	0.5 mm	Cart portable

The response of the active systems is reported in terms of neutron doubles rate, hereafter referred to as doubles rate. For the active systems, the doubles rate is obtained by simulating the active interrogation by AmLi neutrons, while neglecting other potential source terms, such as ^{238}U spontaneous fissions, or (α, n) reactions. This simulation procedure is expected to yield similar results to those obtained during in-field operation. Experimentally, the doubles rate is obtained by subtracting the neutron coincidences passively measured without the interrogating sources from a subsequent active assay. For passive, self-interrogating systems, the doubles and singles rates are net quantities. The net counts are obtained by subtracting the reference doubles rate, only due to ^{238}U spontaneous fissions, from the simulated doubles rate from a given assembly.

All the participating groups calculated the doubles rate as the second moment of the neutron multiplicity distribution derived by using shift-register triggered counting gates [7], [10]. The following

gate times were used: 0.1 μs (UM and INL), 0.512 μs (LLNL), and 200 μs (LANL). The gate intervals were optimized by selecting the width that minimized the relative standard deviation associated with the doubles rate, using the methods outlined by Prasad et al. [7] and Croft et al. [11], for the active systems based on fast and thermal neutron detectors, respectively. The passive neutron systems have such a low totals rate (approx. 300 cps) that the longer gate can be used to provide minimum error.

In the simulated FNCL configuration, cross talk coincidences account for approximately 30% of the total number of coincidences. All the groups but the LLNL team did not discriminate and filter neutron coincidences due to cross talk. The neutron cross talk contribution is expected to be negligibly affected by the assembly activity or by other parameters estimated in this analysis, e.g. sensitivity to burnable poisons. Therefore, discriminating the cross talk would only result in a negative bias in the trend of the doubles rate as a function of ^{235}U and, as a consequence, negligibly affect the ultimately estimated ^{235}U LD.

The simulated detection systems vary by detection material (^3He , ^{10}B , stilbene, organic glass, and EJ-309), type of assay (passive or active), geometry and number of utilized interrogation sources, in the active case. The performance of each system is a convoluted function of these features. Different metrics are used to compare the systems, including the mass defect, i.e. the relative difference of the estimated ^{235}U LD compared to the declared one, the assay penetrability (Section VI.c), and the 3σ test (Section VI.c). Ultimately, these metrics are used to compare the systems in terms of sensitivity to the assembly form factor, sensitivity to potential diversion scenarios and to burnable poisons in the assemblies.

a. Calibration Assemblies

In this effort, the ^{235}U LD of different fuel bundles was estimated through calibration-based assay. Therefore, the first assay stage consisted in creating a calibration curve by simulating the systems' response to nine assayed 17x17 assemblies, with increasing ^{235}U enrichment and LD ranging from 0 to 65 g cm^{-1} . Fig. 32 shows the doubles count rate for these assemblies as a function of ^{235}U LD. The sensitivity of a measurement system is typically given by the ratio between the readout and the measured quantity [12]. Fig. 32 shows that the EJ-309 based FNCL is the most sensitive system. Despite an overall similar light output of the simulated organic scintillation materials (Fig. 19), the highest EJ-309 FNCL response should be ascribed to the detector composition, the detection volume and the system form factor. For example, stilbene exhibits a light output close to EJ-309 (Fig. 19), however, the sensitivity of stilbene-based systems is lower compared to the sensitivity of the EJ-309 based FNCL. This difference is most probably due to the smaller detection volume of the UM-SB system (approximately 3,000 cm^3), compared to the FNCL (approximately 12,000 cm^3), and the pulse post processing procedure of the LLNL-SB system. Unlike other active systems, the doubles rate response of the LLNL-SB system does not include neutron coincidences due to cross-talk [13].

Fig 32 also includes the calibration curves used by each system. Table 11 summarizes the fit functions and calibration coefficients used by each system.

Figure 32. Simulated calibration curves of the systems participating to the Neutron Rodeo Phase II.

Table 11. . Functional fit curves and coefficients used for calibration.

Detection System	Fit function	a	b	c
FNCL	$D = \left(\frac{aL}{1 + bL} \right) + c$	$1.496 \pm 4.94\text{E-}02$	$0.007 \pm 6.44\text{E-}04$	$5.202 \pm 5.50\text{E-}01$
UM-SB	$D = \left(\frac{aL}{1 + bL} \right) + c$	$0.570 \pm 1.42\text{E-}04$	$0.005 \pm 1.54\text{E-}07$	$4.500 \pm 1.94\text{E-}02$
LLNL-SB	$D = \left(\frac{aL}{1 + bL} \right)$	$1.210 \pm 7.57\text{E-}02$	$0.010 \pm 1.85\text{E-}03$	N.A.
INL- organic glass	$D = \left(\frac{aL}{1 + bL} \right)$	$1.740 \pm 4.47\text{E-}02$	$0.012 \pm 9.09\text{E-}04$	N.A.
LANL – He-3	$D = aL + b$	$0.354 \pm 6.70\text{E-}02$	$1.042 \pm 1.72\text{E-}03$	N.A.
LANL – B-10	$D = aL + b$	$0.001 \pm 1.79\text{E-}03$	$0.108 \pm 4.24\text{E-}04$	N.A.

This desirable linear calibration curve of the passive systems is mostly due to intrinsically higher penetrability of the assay, compared to active assays based on external AmLi neutrons.

When comparing the calibration curves of active and passive systems, it should be noted that the latter do not require any external interrogation source. Therefore, it is useful to report the calibration results per unit interrogating neutron for the active systems (Fig. 33), as comparison.

Figure 33. Simulated calibration curves of the systems based on organic scintillators participating to the Neutron Rodeo Phase II.

b. Intact Assemblies

A variety of intact assemblies was simulated to test the systems' sensitivity to assemblies with a ^{235}U LD within the range of calibration, but with a form factor slightly different from the 17x17 calibration assemblies. The simulated mass defects are shown in Fig. 34. A desirable mass defect close to zero is reported by all the systems for the 17x17 assemblies (Fig. 34). These assemblies consist of 264 pins and 25 empty channels like the calibration assemblies, and ^{235}U LD of 41.3 g cm^{-1} and 54.8 g cm^{-1} , in the case of the 17x17A and 17x17B, respectively. Conversely, the 14x14A and 14x14B assemblies encompass 179 and 176 fuel pins and a ^{235}U LD of 37.6 g cm^{-1} and 37.3 g cm^{-1} , respectively. 14x14A and 14x14B also differ for the geometric distribution of the empty channels within the assembly, which are uniformly distributed in the 14x14A case, while arranged in five clusters of four channels each in the 14x14B case. Despite the similar ^{235}U , only two systems, i.e. the LLNL-SB and the SNL organic glass, exhibit a zero mass-defect within one standard deviation for both 14x14A and 14x14B bundles. It should be noted that the side panels of the FNCL were moved close to the 14x14 bundle, to keep the distance from the

bundle side to the front face of the panel consistent with the assay of 17x17 bundles. Fig. 34 also shows that both passive systems under predict the LD ^{235}U in the case of the 14x14 assemblies. This effect is due to the heavy metal mass difference in the four assembly types. The different linear uranium mass loadings were $\sim 909, 1192, 1291$, and 1304 g/cm for the 14x14A, 14x14B, 17x17A, 17x17B, respectively. The neutrons resulting from the spontaneous fission reactions scatter in the fuel assembly, including the mass of the fuel assembly that extends above and below the detectors that acts as reflectors. This scattering increases the detection efficiency for the thermal-neutron detectors. The current IAEA collar software (INCC) has a correction factor called k_4 to correct for the mass difference from the calibration assembly.

Figure 34. Simulated mass defect for the intact assemblies.

As a matter fact, the passive assay based on the singles count rate shows instead a positive mass defect of approximately 15% and 3% for the 14x14A and 14x14B assemblies, respectively. This change in the measured singles rates (per unit fuel mass) is related to the reduced size of the 14x14 fuel assemblies that provides a larger solid angle for the passive neutrons from the ^{234}U to hit the detector slabs. The FNCL shows both a positive and negative bias for the 14x14A and 14x14B, respectively depending on the fuel panel distance of the fuel assembly inside the detector slabs. The fuel-panel distance of the FNCL and the form factor of the passive system can be further optimized to mitigate the assay sensitivity to bundle size.

c. Partial Defect Assemblies

One of the main applications of nuclear collars is the detection of missing or replaced fuel pins in a potential diversion scenario. In this section, we compare the sensitivity of the simulated systems to a diversion of 1, 8, or 40 fuel pins, and their replacement with iron, DU, and lead rods.

The declared LD of PDAs corresponds to the full assembly. Therefore, a low mass defect is desirable because a large deviation from the declared LD allows the operator to identify a fuel diversion. Fig 35-37 show that that one pin, of either iron, DU, or lead, does not yield a significant mass defect for any of the systems. Conversely, all the systems detect a 40-pin replacement, regardless of the material. The passive systems are also able to detect an 8-pin detection, except in the DU replacement case, because in this case the ^{238}U term still contributes to overall signal.

Active systems are generally insensitive to a diversion of eight pins, with the exceptions of the FNCL and the LLNL-SB system when assaying bundles with DU or iron replacements. The consistently lower sensitivity to a fuel replacement with lead, i.e., higher mass defects, is most probably due to the lower neutron capture cross section of lead, compared to DU and iron.

Figure 35. Simulated mass defect for the partial defect assemblies, with 1, 8, and 40 fuel rods replaced by depleted uranium.

Figure 36. Simulated mass defect for the partial defect assemblies, with 1, 8, and 40 fuel rods replaced by lead rods.

Figure 37. Simulated mass defect for the partial defect assemblies, with 1, 8, and 40 fuel rods replaced by iron rods.

In the assay of PDAs using passive systems, the mass defect calculated using the neutron singles rate yields consistently more accurate results (Fig. 38), compared to the doubles rate based assay by the same systems.

Figure 38. Simulated mass defect for the partial defect assemblies, with 1, 8, and 40 fuel rods replaced by lead and iron rods. Mass defects are derived using the passive systems using single and double count rates.

PDAs with a replacement of eight or forty pins were simulated in two different spatial configurations, the first with pin removal from the middle of the assembly and the second, A, with uniform pin removal. The ratio between the mass defect in the two spatial configurations is a metric of

Figure 39. Ratio between simulated mass defects with uniform pin replacement and replacement from the center of the assembly.

the penetrability of the assay. Fig 39 shows that this ratio is close to one for the passive systems, signifying a low dependence on geometry distribution of replaced fuel pins due to the distributed nature of the interrogating source in the self-interrogating passive assay.

One metric of system sensitivity to the diversion of fuel pins is the so-called 3σ test. This test consists of determining whether the declared ^{235}U LD is within $\pm 3\sigma$ of the assayed value. Table 12 shows the results of this analysis for each PDA. In Table 12 'N' means that declared ^{235}U LD is not within $\pm 3\sigma$ of the assayed value, i.e., a fuel diversion was successfully detected.

Table 12. Results of the 3σ test for the partial defect assemblies.

Detection System	DU 1	DU 8	DU 8a	DU 40	DU 40a	Fe 1	Fe 8	Fe 8a	Fe 40	Fe 40a	Pb 1	Pb 8	Pb 8a	Pb 40	Pb 40a
FNCL	Y	Y	Y	N	N	Y	Y	Y	N	N	Y	Y	Y	N	N
UM-SB	Y	Y	Y	N	N	Y	Y	Y	N	N	Y	Y	Y	N	N
LLNL-SB	Y	Y	Y	N	N	Y	Y	Y	N	N	Y	Y	Y	N	N
INL- organic glass	Y	Y	Y	N	Y	Y	Y	Y	N	N	Y	Y	Y	Y	Y
LANL – He-3	Y	Y	Y	Y	Y	Y	N	N	Y	Y	Y	N	N	Y	Y
LANL – B-10	Y	Y	Y	Y	Y	Y	N	N	N	N	Y	N	N	N	N

d. Poisoned Assemblies

One of the most critical applications of neutron collars is their use in assaying fuel assemblies loaded with burnable poisons. An increasing number of fuel types incorporate fuel pins enriched in an

Figure 40. Simulated mass defect for the burnable poisons assemblies.

isotope with a high cross section for thermal neutrons, with the objective of controlling the core reactivity at the first stage of reactor operation. These poisons are eventually transmuted, “burned”, during reactor operation, and have a negligible effect on the neutron core dynamics for burnups higher than 10 GWd/tU. In the second Neutron Rodeo phase, gadolinium enriched fuel assemblies were simulated. The number of poisoned pins was 4 and 24, and the concentration of Gd was either 6% or 10%, for each type, adding up to four different burnable poisoned assemblies. The presence of a cadmium liner is particularly important in this assay because it effectively filters low energy neutrons, below the Cd cutoff energy of 0.5 eV, from entering the assembly after being scattered inside the detectors or by materials surrounding the fuel bundle. The Cd liner minimizes the thermal portion of the neutron spectrum and mitigates the effect of Gd capture reactions on the neutron population inside the assembly. The use of the cadmium liner, referred to as fast-mode collar operation in Neutron Rodeo Phase I, in principle allows the application of a calibration curve obtained for un-poisoned assemblies to estimate the ^{235}U LD of poisoned ones. Fig. 40 shows the mass defect in the four cases. In this type of assay, the smaller the mass defect for a given fuel design, the less sensitive the system is to burnable poisons. Therefore, a zero mass-defect is desirable.

With the exception of the FNCL, active systems simulate a mass defect within 1σ of the desired value when 4 pins are enriched with Gd, either at 6% or 10% Gd concentrations. The smaller uncertainty due to the higher sensitivity of the FNCL adversely affects its sensitivity to poisoned fuel. In the case of a 24-pins replacement, all the systems detect a statistically significant diversion ($> 1\sigma$) from the zero mass-defect. In passive systems, using the singles count rate, instead of doubles count rates, is advantageous (Fig. 41).

Figure 41. Simulated mass defect for the burnable poisons assemblies. Mass defects are reported for the passive systems using single and double count rates.

It is also interesting to discriminate the statistic, systematic and calibration contributions to the combined uncertainty [14]. Figure 42 shows that the assumed systematic contribution of 2% is the most relevant one for active systems (Figures 42.a, 42.b, 42.c, 42.f). The predominance of the systematic uncertainty over the statistic and calibration contributions is not surprising given the short coincidence time gates used in active systems and their high sensitivity. Among the different fast systems, an overall lower uncertainty due to the calibration may be noticed for the FNCL and UM stilbene, compared to INL and LLNL collars, which is probably due to the different functional forms used by the first two (Eq. 2), compared to the standard one (Table 11), used by the INL and LLNL systems.

Figure 42. Uncertainties associated with the estimated ^{235}U linear density by (a) UM stilbene collar, (b) LLNL stilbene collar, (c) INL FNCL, (d) LANL He-3 system, (e) LANL B-10 system, and (f) FNCL.

Conversely, in the passive systems the statistic uncertainty plays an important role and is on average higher compared to both the systematic and calibration-related uncertainties. This leaves further room for improvement for these systems to perform longer assay times or possibly using more detectors to reduce the statistic uncertainty contribution.

VII. Weight and Cost estimates

The groups participating in the Neutron Rodeo Phase II also estimated the cost, dimensions, and weight of an instrument potentially deployed in the field. Those estimates are reported in Table 13. The estimated cost for all the systems also includes the electronics, but not the data acquisition workstation (e.g., a desktop/laptop). Additionally, for the active systems, the cost of one or two AmLi interrogating sources should be included. In Table 13, one may notice that the detectors are a fraction of the total system cost that ranges from 40% to almost 90%. This fraction increases for detection technologies not

commercially available or manufactured in custom sizes. It is anticipated that the production cost of new promising materials, such as solution-grown stilbene crystal and organic glass, will continue to decrease with an increasing demand. The cost of He-3 based system strongly depends on the He-3 supplier: an overall detector cost of \$64,000 is foreseen if the gas is part of the NNSA allocated reservoir, while it increases to \$114,000 if commercial gas is used.

Table 13. Estimated cost and physical footprint of instruments based on the detector materials considered in the Rodeo Phase II. IAEA FNCL size does not include the mechanical cart.

Group	Detector Material	Total Active Volume for Full System	Full System Dimensions (L cm x W cm x H cm)	Estimated Cost (\$)	Detector Cost Fraction (approx. %)	Estimated Weight (kg)
IAEA (Benchmark FNCL)	EJ-309 liquid organic	12,000 cm ³	57 x 51 x 82	NA	40	275
LLNL	Trans-stilbene	12,356 cm ³	63 x 50 x 90	300,000	88	150
UM	Trans-stilbene	3,089 cm ³	40 x 40 x 76	150,000	60	100
SNL/INL	Small-molecule organic glass	12,000 cm ³	57 x 51 x 82	105,000 - 120,000	30	275
LANL1	He-3 tubes embedded in polyethylene	73,440 cm ³ including polyethylene bank	44 x 44 x 51	149,000-199,000	42-57	60
LANL2	Boron-10 cells embedded in polyethylene	129,000 cm ³ including polyethylene bank	47 x 47 x 55	155,000	45	130

VIII. Summary and Conclusions

In the Phase II of the Advanced Neutron Detection Technology Rodeo, five different fresh-fuel safeguards systems were simulated, and their performance was compared to a model of the IAEA FNCL. All the systems used the same methodology to estimate the ²³⁵U linear density of inspected fresh fuel assemblies, which consisted of measuring the neutron doubles rate and using a calibration-based procedure to estimate the fuel ²³⁵U LD.

We simulated four different versions of the FNCL, each based on a different organic scintillator: stilbene, EJ-309, EJ-276, and organic glass, to perform a direct comparison of detector materials, while keeping the same detection volume and form factor. All the simulation and post-processing parameters were kept consistent among the systems. As expected, the systems showed similar performance, with the highest sensitivity shown by the EJ-309 based FNCL, thanks to its high neutron light output and

hydrogen concentration. The EJ-309-based FNCL is also the most sensitive to the presence of burnable poisons in the inspected assemblies, on average. With respect to unpoisoned fuel assemblies, regardless of the material, an average drop of the neutron doubles signal of 2.2% and 8.4% results from the insertion of 6wt% and 10 wt% Gd-loaded pins, respectively. The stability over-time of newly developed materials such as EJ276 and organic glass still needs to be demonstrated, however, they are safer to operate compared to liquids.

Each research team participating in the Phase II of the Advanced Neutron Detection Technology Rodeo designed a realistic system with various detector types and assay techniques. All the results from each group were then compared directly with the results from a benchmark simulation of the FNCL. The five systems can be classified into two main groups, according to the rationale used to induce fission on ^{235}U . The first group relied on external AmLi sources to actively interrogate the fresh fuel and detected fast neutrons using fast-timing, PSD and spectroscopy capable organic scintillators. The second group implemented a self-interrogation model where the induced fissions on ^{235}U are triggered by neutrons emitted by ^{238}U spontaneous fission neutrons. In this case, emitted neutrons are moderated, and detectors sensitive to thermal neutrons are used to exploit their high intrinsic efficiency in this energy range.

The performance of each system was compared to the FNCL in terms of sensitivity to fissile mass and fuel form factor, assay penetrability, sensitivity to burnable poisons and fuel pins' diversions.

While the FNCL exhibits the highest sensitivity to fissile mass, all the systems except for the LLNL collar and INL FNCL estimated a ^{235}U LD ($> 1\sigma$) different than the declared LD of 14x14 assemblies, smaller than the 17x17 ones used for calibration. This result is probably due to the type of calibration fit function adopted by these two systems.

Active systems are generally insensitive to a diversion of eight or fewer (one) pins, with the exceptions of the FNCL and the LLNL-SB system when assaying bundles with 8-pin DU or iron replacements. Fuel replacement with lead yields consistently higher mass defects. This effect is most probably due to the lower neutron capture cross section of lead, compared to DU and iron, and makes a lead diversion intrinsically harder to detect, compared to a DU or iron diversion.

PDAs with a replacement of eight or forty pins were simulated in two different spatial configurations, the first with pin removal from the middle of the assembly and the second with uniform pin removal. The system relative response to these assemblies can be used to compare the assay penetrability. As expected, passive systems exhibit an optimal penetrability, while active systems show large diversions depending on the distribution of the replaced pins. Penetrability can be improved in active systems by using higher energy interrogation sources, e.g., D-D generators.

In response to burnable poisons, active systems exhibit mass defect within 1σ of the desired value when 4 pins are enriched with Gd, either at 6% or 10% Gd concentrations, with the exception of the FNCL. The smaller uncertainty due to the higher efficiency of the FNCL makes it more sensitive to the presence of poisoned fuel, compared to other systems. In the case of a 24-pins replacement, all the

systems detect a statistically significant diversion ($> 1\sigma$) from the desirable zero mass defect of a system virtually insensitive to burnable poisons.

The NDA results for the self-interrogation mode systems (i.e., LANL ^3He and LANL ^{10}B) overall improved the sensitivity to PDAs relative to the active-interrogation mode systems, where the self-interrogation systems were able to detect a removal of eight metal pins with $> 2\sigma$ confidence. The self-interrogation mode systems can also use the neutron singles count rates due to the absence of interrogation sources; the NDA results using the singles and doubles count rates were also compared. Overall, the singles and doubles count rates performed comparably for all types of assemblies. The use of singles count rate provides improved statistics, but it is also more affected by non-negligible singles background counts in practical settings, compared to doubles count rates.

Emerging detector technologies and a new assay modality were compared to the FNCL. From the reported results, the collar technology should be selected according to the type of assay that one needs to perform. FNCL and the LLNL collar exhibit the best performance in detecting PDAs. Should the pin be replaced with heavy metals, passive systems would be the most sensitive to the diversion. Conversely, relatively low efficiency systems like the UM stilbene collar, INL organic glass FNCL, or B-10 based collar could be used to minimize the assay sensitivity to burnable poisons. Safeguards systems based on organic scintillators also allow the detection of multiplets higher than the second order, whose use should be further investigated to be able to estimate other relevant information of the fuel bundle, such as the multiplication and overall neutron source strength.

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IX. References

- [1] S. A. Pozzi *et al.*, “MCNPX-PoliMi for nuclear nonproliferation applications,” *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 694, pp. 119–125, Dec. 2012.
- [2] E. C. Miller, S. D. Clarke, M. Flaska, S. A. Pozzi, and E. Padovani, “MCNPX-PoliMi post-processing algorithm for detector response simulations,” *J. Nucl. Mater. Manag.*, vol. 40, no. 2, pp. 34–41, 2012.
- [3] A. Enqvist, C. C. Lawrence, B. M. Wieger, S. A. Pozzi, and T. N. Massey, “Neutron light output response and resolution functions in EJ-309 liquid scintillation detectors,” *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 715, pp. 79–86, Jul. 2013.
- [4] M. Monterial, P. Marleau, and S. Pozzi, “Characterizing subcritical assemblies with time of flight fixed by energy estimation distributions,” *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 888, pp. 240–249, 2018.
- [5] G. F. Knoll, *Radiation detection and measurement*, Fourth. Hoboken, N.J. : John Wiley, c2010., 2010.
- [6] A. Belian *et al.*, “Advanced Neutron Detection Technology Rodeo,” *Proc. or ESARDA 39th Annu. Meet. Dusseldorf, Ger.*, 2017.
- [7] M. K. Prasad, N. J. Snyderman, and J. M. Verbeke, “Analytical error bars and RSD for neutron multiplicity counting,” *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 903, pp. 25–31, 2018.
- [8] J. B. Birks, *The Theory and Practice of Scintillation Counting*. Pergamon Press, 1964.
- [9] M. M. Bourne, S. D. Clarke, N. Adamowicz, S. A. Pozzi, N. Zaitseva, and L. Carman, “Neutron detection in a high-gamma field using solution-grown stilbene,” *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 806, pp. 348–355, 2016.
- [10] W. Hage and D. M. Cifarelli, “Correlation analysis with neutron count distributions in randomly or signal triggered time intervals of special fissile materials,” *Nucl. Sci. Eng.*, vol. 89, no. 2, 1985.
- [11] S. Croft, D. Henzlova, A. Favalli, D. K. Hauck, and P. A. Santi, “The optimum choice of gate width for neutron coincidence counting,” *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 764, pp. 322–329, Nov. 2014.
- [12] A. D. McNaught and D. Wilkinson, *IUPAC. Compendium of Chemical Terminology, 2nd ed.* Oxford, 2006.
- [13] J. M. Verbeke, M. K. Prasad, and N. J. Snyderman, “Neutron crosstalk between liquid scintillators,” *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 794, pp. 127–140, Sep. 2015.
- [14] ISO IUPAC OIML BIPM IEC IFCC ILAC, “JCGM 100:2008 Evaluation of measurement data — Guide to the expression of uncertainty in meas.” Joint Committee for Guides in Metrology, 2008.

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Appendix A

We calculated the uncertainty σ_L of the linear density, L , (Eq 12) by propagating the uncertainties associated with the simulated doubles rate D and with each coefficient of the calibration model (Eq 6-7): a , b , and c . The single contributions are detailed in Eq 8 – 11 .

$$D = a \frac{L}{1+bL} + c \quad (6)$$

$$L = \frac{D-c}{a+b(c-D)} \quad (7)$$

$$\frac{\delta L}{\delta D} = \frac{a}{(a+b(c-D))^2} \quad (8)$$

$$\frac{\delta L}{\delta a} = \frac{c-D}{(a+b(c-D))^2} \quad (9)$$

$$\frac{\delta L}{\delta b} = \frac{(c-D)^2}{(a+b(c-D))^2} \quad (10)$$

$$\frac{\delta L}{\delta c} = -\frac{a}{(a+b(c-D))^2} \quad (11)$$

$$\sigma_L = \sqrt{\left(\left(\frac{\delta L}{\delta D}\right)^2 \sigma_D^2 + \left(\frac{\delta L}{\delta a}\right)^2 \sigma_a^2 + \left(\frac{\delta L}{\delta b}\right)^2 \sigma_b^2 + \left(\frac{\delta L}{\delta c}\right)^2 \sigma_c^2 + 2\left(\frac{\delta L}{\delta a}\right) \frac{\delta L}{\delta c} \sigma_{ac} + 2\left(\frac{\delta L}{\delta b}\right) \frac{\delta L}{\delta a} \sigma_{ab} + 2\left(\frac{\delta L}{\delta b}\right) \frac{\delta L}{\delta c} \sigma_{bc}} \quad (12)$$

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