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Fast-Neutron Multiplicity Counter for the Detection of Diversion Scenarios

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ABSTRACT

We demonstrated the application of a fast-neutron multiplicity counting (FNMC) system to detect small variations in an assembly of uranium-oxide pins. The current version of the FNMC includes eight liquid organic scintillators (EJ-309 7.62 cm diameter x 7.62 cm) and eight crystal organic scintillators (stilbene 5.08 cm diameter x 5.08 cm) in a checkerboard assembly. We measured time-correlated fast neutrons, emitted by an assembly of 32 uranium-oxide pins, using two Am-Li sources to induce fissions in the sample. Each pin is a cylinder of approximately 1 cm diameter and 10 cm length, with a mass of 75.52 g and 16.37 wt% enrichment. We removed up to 16 pins from a sample consisting of 32 pins, and determined the minimum number of pins that resulted in a statistically significant decrease of neutron coincidence counts. All the measurements were performed at the Zero Power Physics Reactor facility of Idaho National Laboratory. We found that a diversion of the four innermost pins could be detected by the system in 5 minutes with 95% confidence. Fast-neutron measurements have several potential advantages compared to thermal and epithermal neutron counters the most important being a shorter die-away time and therefore reduced spurious coincidences due to accidental counts. This advantage may result in a higher sensitivity in timely detecting diversion of nuclear material from peaceful uses.

I. Introduction and Motivation

International nuclear safeguards are implemented to verify that the commitments made by the States under the safeguards agreements are fulfilled. Of the many different objectives of international safeguards, one particular scenario of interest is the diversion of declared nuclear material from a safeguarded facility [1]. Specifically, improving on the speed and accuracy of detecting diverted material from fuel assemblies can have significant benefits on the procedural and costs aspects of inspections. The standard system currently used by the International Atomic and Energy Agency (IAEA) involves an array of capture-based ^{3}He proportional counter embedded in polyethylene with an Am-Li interrogative neutron source; this system is known as the Uranium Neutron Collar (UNCL). The thermalized Am-Li neutrons induce fissions within the fuel assembly, which initiates fission chains. The detection of the multiplets of neutrons emitted from these chains allows for a quantitative characterization of the assembly. Recent research efforts have investigated alternate detector materials to improve on the current capabilities of the UNCL. A promising new technology is the use of scatter-based organic scintillators to detect unmoderated fast fission neutrons emitted directly from the multiplying system; these systems are referred to as Fast-Neutron Multiplicity Counters (FNMC).

Scatter-based organic scintillators (e.g. EJ-309 liquid, BC-501 plastic, stilbene crystal, etc.) detect fast neutrons (~ > 0.5 MeV) by collecting the light created from the recoil proton within the scintillator active volume. Therefore, no moderation of the neutrons prior to detection is necessary. Furthermore, organic scintillators exhibit relatively fast response times (on the order of tens of nanoseconds) and good timing resolution (~ 1 nanosecond at FWHM) that is ideal for time-correlation analysis of detected counts. The advantages of an FNMC system are [2-3]:

1. Fast system die-away time: as there is no need for moderating material between the assembly and the detectors, there is no "neutron slowing-down time"

2. Short time gates for correlating detected counts (on the order of 10s to 100s of nanoseconds) due to the fast response time and timing resolution: this leads to a much fewer random coincidences.

3. Lower overall counting uncertainty due to the timescale of FNMC systems: although FNMC systems may have lower detection efficiency relative to UNCL systems, the time scale of FNMC systems means that multiplicity counts can be acquired many more times for a fixed measurement time. This can reduce acquisition times and simultaneously reduce measurement uncertainties for a fixed acquisition time.

II. Experimental Setup and Data Acquisition

a. FNMC System Design

A prototype FNMC system consisting of eight 7.62Ø cm x 7.62 cm EJ-309 liquid organic and eight 5.08Ø cm x 5.08 cm stilbene crystal scintillators has been designed and built to test the sensitivity to diversion scenarios. The detectors are arranged in a well-type geometry, and were placed in alternating positions to maintain symmetry. A CAEN V1730 waveform digitizer (500 MHz sampling rate, 14-bit resolution, 2V dynamic range, 16 channels) was used digitize and acquire pulses from all sixteen detectors. Two Am-Li interrogative neutron sources (50,000 n/s) were placed above and below the FNMC system; the sources were embedded in polyethylene to thermalize the emitted neutrons. An image and schematic of the FNMC system is shown in Figure 1.

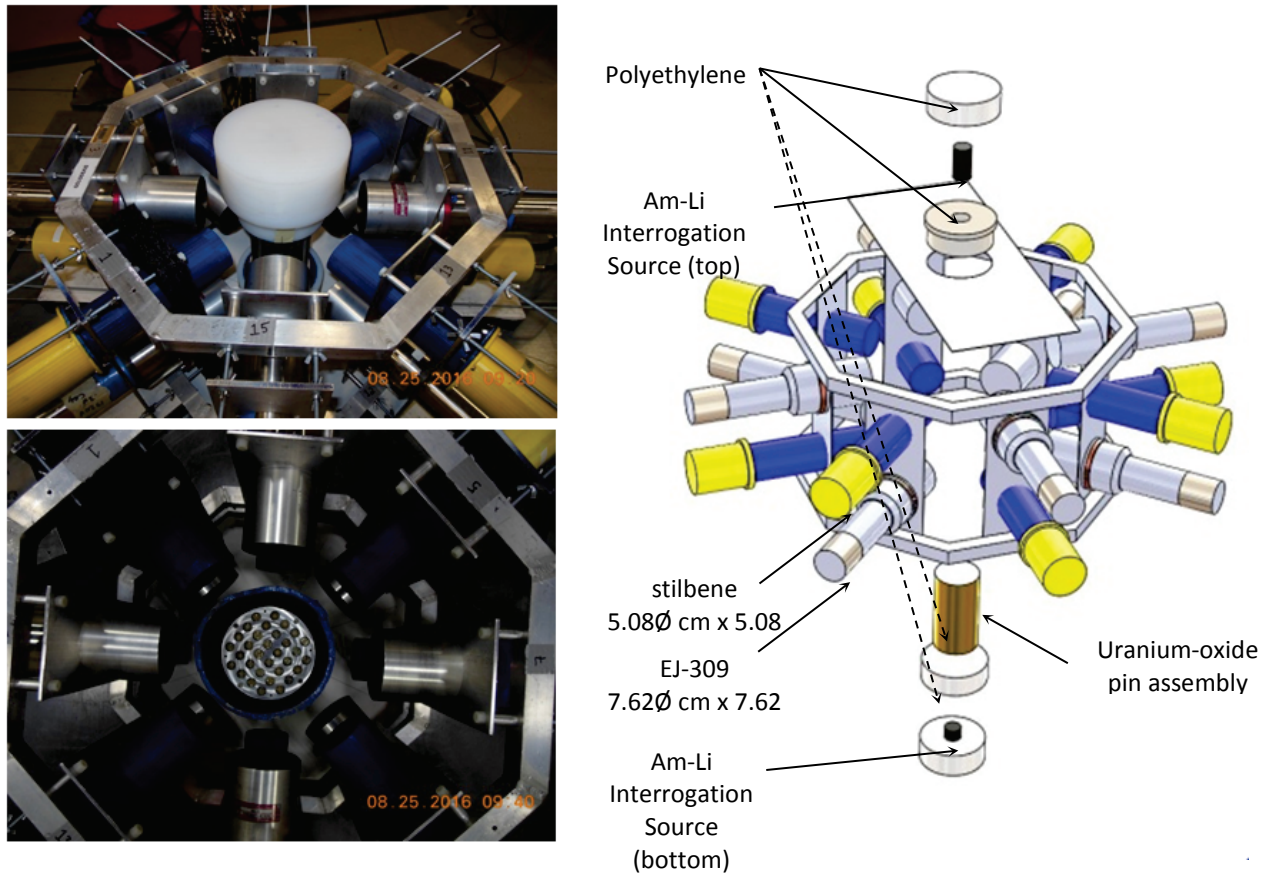


Figure 1. An image of the FNMC system during acquisition (top-left) and an image of the full uranium-oxide pin assembly (bottom-left). A schematic diagram (right) shows the components of the FNMC system for active-mode assay.

b. Uranium-oxide Pins

The interrogated uranium oxide pins had a ^{235}U enrichment of 16.37 % and a mass of 79.52 g; details can be seen in Table 1. The full assembly consisted of 32 pins arranged in the configuration shown in Figure 2. Once the full assembly was measured and characterized, pins were removed from the full assembly sequentially and the partial assemblies were again interrogated and measured. Details of all assembly scenarios are summarized in Table 2. To mitigate instances of pulse pile-up, lead shielding was placed around the assembly for all scenarios.

Table 2. Details of an individual uranium-oxide pin from the ZPPR facility at INL.

Total Mass (Clad + Fuel)	Clad Mass	Fuel Mass (Uranium + Oxygen)	Uranium Mass (16.37% enriched)	Length	Diameter
102.75 g	12.46 g	90.29 g (79.52 g U) (10.72 g O)	79.52 g (13.02 g ^{235}U) (66.50 g ^{238}U)	15.24 cm	0.95 cm

Table 1a. Configuration of the full assembly.

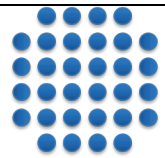
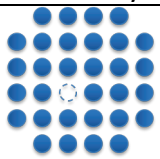
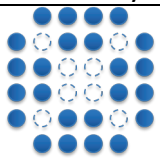
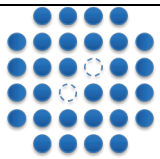
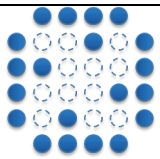
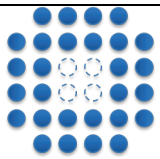
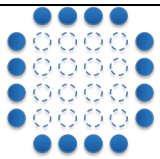
Full Assembly	Pins	Uranium Mass	Defect
	32	2544.64 g	**0 %**

Table 1b. Configurations of the diverted assemblies.

Diverted Assembly	Pins	Uranium Mass	Defect	Diverted Assembly	Pins	Uranium Mass	Defect
 1 removed	31	2465.12 g	**-3.13 %**	 8 removed	24	1908.48 g	**-25.00 %**
 2 removed	30	2385.60 g	**-6.25 %**	 12 removed	20	1590.40 g	**-37.50 %**
 4 removed	28	2226.56 g	**-12.50 %**	 16 removed	16	1272.32 g	**-50.00 %**

c. Data Acquisition

The full waveforms of all detections in each of the organic scintillators were acquired. The organic scintillators used in the FNMC system are sensitive to both neutrons and gamma rays; therefore, pulse shape discrimination (PSD) techniques were implemented for particle identification. Once particles were sorted by particle type, the neutron detections from all detectors were collected and time-sorted. The time-sorted data was used to extract the neutron doubles count rate for all configurations using a signal-triggered time-correlation analysis.

III. Data Analysis

The neutron doubles count rate (**D**) and the associated counting uncertainty (σ_D) was calculated for all configurations for acquisition times of 60, 180, and 300 seconds. The sensitivity for detecting diverted pins were compared for various the three acquisition times by comparing the relative deviation in **D** from the full assembly and implementing a 2σ test for the diverted assemblies. We define the relative deviation in **D** (RD) from the full assembly as

$$\mathrm{RD} = \frac{\boldsymbol{D_{Full}} - \boldsymbol{D_{Diverted}}}{\boldsymbol{D_{Full}}}, \tag{1}$$

and the associated uncertainty in RD (σ_{RD})

$$\sigma_{RD} = \sqrt{\left(\frac{\delta RD}{\delta D_{Full}}\right)^2 \sigma_{D_{Full}}^2 + \left(\frac{\delta RD}{\delta D_{Diverted}}\right)^2 \sigma_{D_{Diverted}}^2}\ . \tag{2}$$

Once RD and σ_{RD} are calculated for each diverted assembly, we calculated the number of standard deviations in the deviations of D using

$$\#\sigma = \frac{RD}{\sigma_{RD}}\ . \tag{3}$$

If #σs are greater than two for a given diverted assembly, then the system was able to detect the diversion scenario with > 95% confidence.

IV. Results & Discussion

Figure 3-5 shows the relative deviations in **D** for all diverted assemblies for a 60, 180, and 300 second acquisition time, respectively.

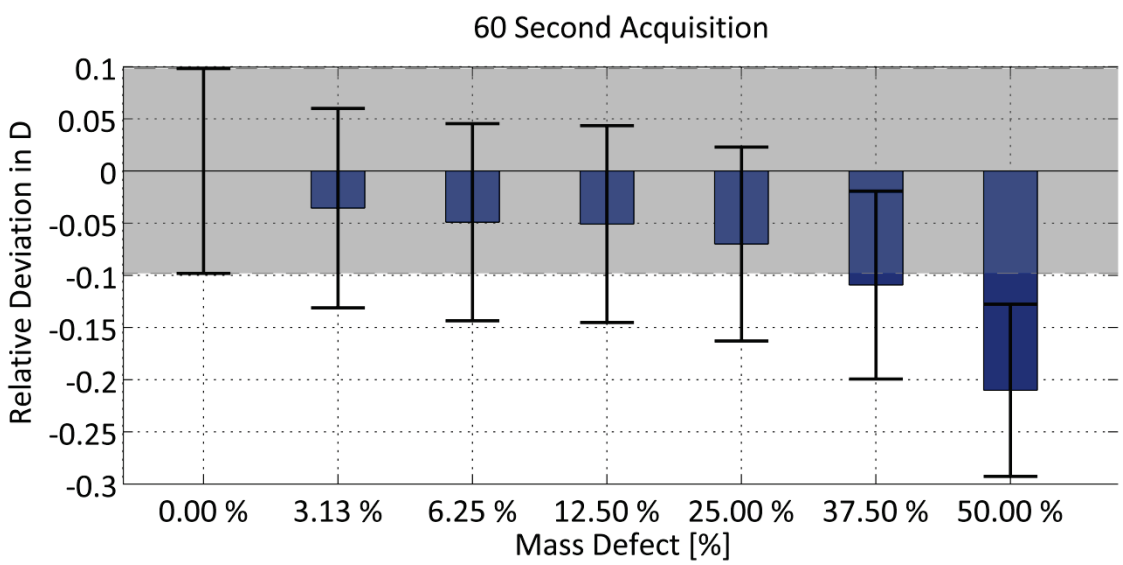


Figure 3. Plot shows the relative deviation in **D** of the diverted assemblies from the full assembly for a 60 second acquisition. The gray shaded region represents the 2σ interval of the measured **D** of the full assembly, and the error bars represent the 2σ uncertainty in the measured **D** for all assemblies.

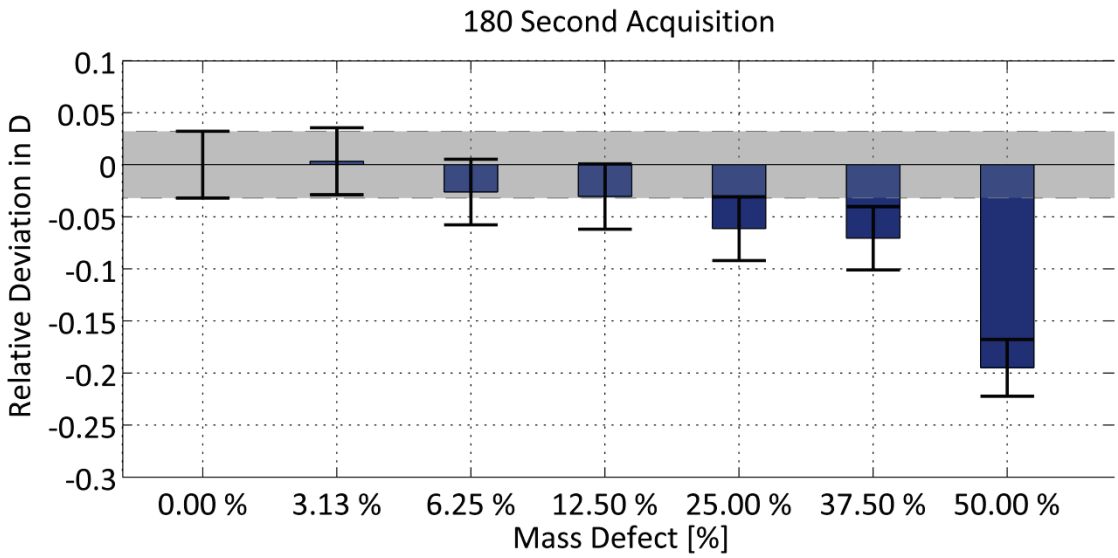


Figure 4. Plot shows the relative deviation in **D** of the diverted assemblies from the full assembly for a 180 second acquisition. The gray shaded region represents the 2σ interval of the measured **D** of the full assembly, and the error bars represent the 2σ uncertainty in the measured **D** for all assemblies.

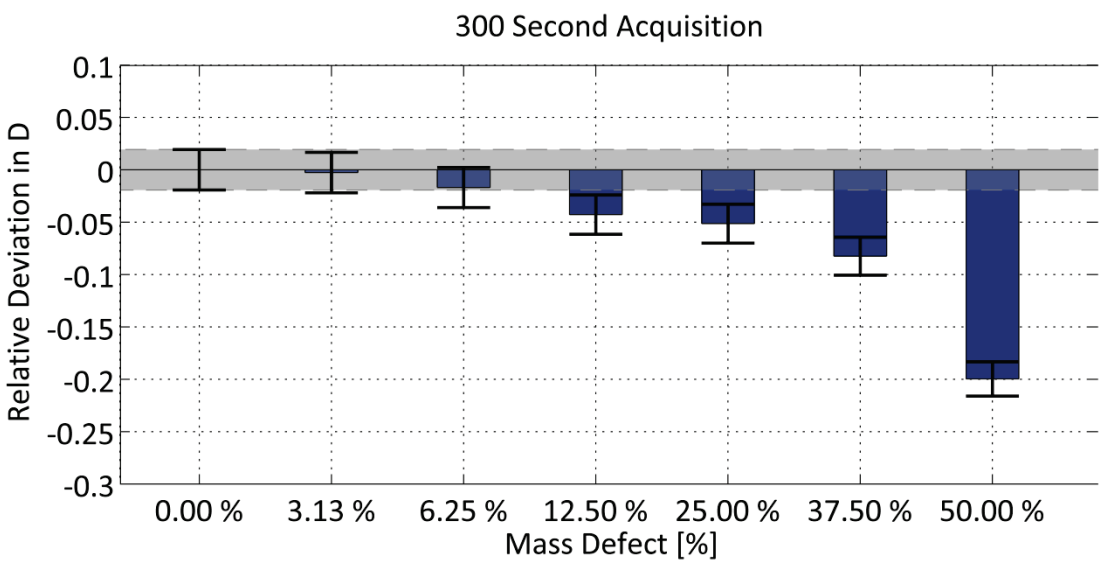


Figure 5. Plot shows the relative deviation in **D** of the diverted assemblies from the full assembly for a300 second acquisition. The gray shaded region represents the 2σ interval of the measured **D** of the full assembly, and the error bars represent the 2σ uncertainty in the measured **D** for all assemblies.

Table 2-4 summarizes the results for the diverted assemblies for the 60, 180, and 300 second acquisitions.

Table 2. A summary of the FNMC sensitivity of detecting diverted pins in a 60 second acquisition.

Number of pins removed	Mass defect [%]	Neutron doubles rate **D** [cps]	Uncertainty in **D** σ_D	Relative deviation (RD) in **D** from full assembly	Uncertainty in RD σ_{RD}	Number of standard deviations from full assembly (#σs)	Diversion detected with > 95% confidence (>2σs)?
0	0.00 %	17.32	0.69	0	0.098	0	No
1	3.13 %	16.70	0.68	-0.036	0.096	0.373	No
2	6.25 %	16.47	0.68	-0.049	0.095	0.520	No
4	12.50 %	16.44	0.69	-0.051	0.094	0.540	No
8	25.00 %	16.11	0.67	-0.070	0.093	0.754	No
12	37.50 %	15.43	0.66	-0.109	0.090	1.215	No
16	**50.00 %**	**13.68**	**0.62**	**-0.210**	**0.083**	**2.548**	**Yes**

Table 3. A summary of the FNMC sensitivity of detecting detect diverted pins in a 180 second acquisition.

Number of pins removed	Mass defect [%]	Neutron doubles rate **D** [cps]	Uncertainty in **D** σ_D	Relative deviation (RD) in **D** from full assembly	Uncertainty in RD σ_{RD}	Number of standard deviations from full assembly (#σs)	Diversion detected with > 95% confidence (>2σs)?
0	0.00 %	16.67	0.23	0	0.032	0	No
1	3.13 %	16.73	0.23	0.003	0.032	0.103	No
2	6.25 %	16.23	0.22	-0.026	0.031	0.837	No
4	12.50 %	16.16	0.22	-0.031	0.031	0.976	No
8	**25.00 %**	**15.65**	**0.22**	**-0.061**	**0.031**	**2.01**	**Yes**
12	**37.50 %**	**15.50**	**0.22**	**-0.071**	**0.030**	**2.32**	**Yes**
16	**50.00 %**	**13.42**	**0.20**	**-0.195**	**0.027**	**7.13**	**Yes**

Table 4. A summary of the FNMC sensitivity of detecting detect diverted pins in a 300 second acquisition.

Number of pins removed	Mass defect [%]	Neutron doubles rate **D** [cps]	Uncertainty in **D** σ_D	Relative deviation (RD) in **D** from full assembly	Uncertainty in RD σ_{RD}	Number of standard deviations from full assembly (#σs)	Diversion detected with > 95% confidence (>2σs)?
0	0.00 %	16.84	0.14	0	0.019	0	No
1	3.13 %	16.79	0.14	-0.003	0.019	0.142	No
2	6.25 %	16.55	0.14	-0.017	0.019	0.888	No
4	**12.50 %**	**16.12**	**0.13**	**-0.043**	**0.019**	**2.289**	**Yes**
8	**25.00 %**	**15.97**	**0.13**	**-0.051**	**0.019**	**2.765**	**Yes**
12	**37.50 %**	**15.45**	**0.13**	**-0.083**	**0.018**	**4.549**	**Yes**
16	**50.00 %**	**13.48**	**0.12**	**-0.200**	**0.016**	**12.159**	**Yes**

For a 60 second acquisition of the interrogated uranium pin assemblies, the FNMC system was only able to distinguish a 50 % mass diversion as indicated by the results in Figure X and Table 2. As the acquisition time is increased to 180 seconds, the FNMC system was discern a mass diversion of 25 %. For a 300 second acquisition, the system was sensitive to a 12.5 % mass diversion.

V. Conclusions

A fast-neutron multiplicity counting system was used to experimental determine the system sensitivity of detecting diversion scenarios. The system consisted of eight 7.62Ø cm x 7.62 cm EJ-309 liquid organic and eight 5.08Ø cm x 5.08 cm stilbene crystal scintillators. Uranium-oxide pins (16.57 % ^{235}U enriched, 79.52 g per pin) were arranged in various diverted configurations and interrogated with two Am-Li neutron sources to induce fissions; the emitted fast fission neutrons and gamma-ray pulses were acquired. Pulse shape discrimination techniques were used to extract the neutron pulses, and the neutron doubles rates were calculated. The neutron doubles rates of the diverted configurations were compared directly with the doubles rate from the full assembly. To characterize the sensitivity of the system to detect deviations from the full assembly, the number of standard deviations was calculated and a 95% confidence criterion was implemented. Results show that the fast-neutron multiplicity counting system was able to identify a -12.5 % mass diversion scenario in a 300 second acquisition time with > 95%

confidence. This shows promise in the potential to improve the ability for timely detection of diversion scenarios in safeguarded facilities.

VI. References

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3. D.L. CHICHESTER, S.J. THOMPSON, M.T. KINLAW, J.T. JOHNSON, J.L. DOLAN, M. FLASKA, S.A. POZZI "Statistical Estimation of the Performance of a Fast-neutron Multiplicity system for nuclear material accountancy" *Nuclear Instruments and Methods in Physics Research A.,* **784,** 448 (2015)