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^{235}U Determination using an In-Beam Delayed Neutron Counting Technique at the NRU Reactor

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INTRODUCTION

Canadian Nuclear Laboratories (CNL) and the Royal Military College of Canada (RMC) have independently developed delayed neutron counting systems. These systems were built to expand non-destructive assay capabilities available to the Canadian government for detecting and identifying special nuclear materials in unknown samples. In delayed neutron counting, samples are exposed to an interrogation source (in this case, neutrons) which induces fission in any special nuclear material present. Accompanying prompt neutrons and fission products are delayed neutrons, which are released up to several minutes after the fission process. The magnitude and temporal behavior of the delayed neutrons released is the dependent on the isotope undergoing fission. Therefore, by measuring the delayed neutron magnitudes with respect to time, this technique can detect, and identify special nuclear material content.

The delayed neutron re-interrogation technique creates a steady state of delayed neutrons by the repeated exposure of fissionable content to a neutron interrogation source. It offers several advantages, namely improved statistics for many interrogations. This technique was used by Myers *et al* [3] to determine bulk sample enrichments using a 14 MeV neutron source. They determined the ^{235}U enrichment for 10 bulk samples with errors ranging from 1 – 6 % (weight %).

This paper describes a collaborative effort that saw RMC's delayed neutron and gamma counting apparatus transported to CNL for use in the neutron beamline at the National Research Universal (NRU) reactor. Samples containing mg quantities of fissile material were re-interrogated, and their delayed neutron emissions measured. This collaboration offers significant advantages to previous delayed neutron research at both CNL [1] and RMC [2].

The size, sensitivity, and arrangement of RMC's ^{3}He detectors facilitate a detection efficiency approaching 100 times the previous arrangement at CNL [1]. However the pneumatic system used to send samples from the reactor at RMC to the counting arrangement necessitated a delay in data acquisition. The minimum time required to accommodate sample transfer was 3 *s* and valuable early time delayed neutron emissions were not recorded [4]. This problem is resolved in the CNL set up as the sample remains stationary and its exposure to the neutron beam occurs *in situ*. This results in a drastically reduced delay between the termination of irradiation and the commencement of data collection.

Although the flux of the neutron beamline at CNL (10^{7} $cm^{-2}s^{-1}$) was significantly lower than that of RMC's reactor (10^{12} $cm^{-2}s^{-1}$) it was compensated in the new set up by an increase in fissile mass analyzed and repeated irradiation and counting cycles for each sample using the re-interrogation technique.

Finally, the lower count rates and improved electronics at CNL reduce dead time effects, which distort measurements at RMC and demand data correction. Thus facile data analysis enables more accurate temporal measurements, which are necessary when discerning between different fissile isotopes and their mixtures via delayed neutron counting.

This work details the determination of ^{235}U content in enriched uranium via the assay of in-beam delayed neutron magnitudes and temporal behavior.

EXPERIMENTAL

Three samples of UO_2 were prepared with natural, 1.7, and 2.7 % ^{235}U isotopic abundances to assess the performance of new arrangement. Sample capsules are shown in Figure 1. The cylindrical samples were 10-11 mm in diameter and 10-14 mm in length with total masses ranging from 10.39 to 13.63 g. The capsules were designed specifically to match the requirements of the RMC detector. These samples were placed in the center of six ^{3}He detectors (RSP4-1613-202, GE Energy) which were embedded in paraffin, as described in reference 3. Samples were placed directly in-line with the neutron beam from NRU, Figure 2.

Measurements were performed at the E3 beamline at NRU. A monochromatic beam of nominally 14.6 meV neutrons was provided by diffraction from a Pyrolytic Graphite monochromator. The RMC detector was placed on the usual sample table with both the sample tube and the γ port horizontal as illustrated in Figure 2. Beam collimating components provided a beam 25 mm wide and 50 mm tall.

The support system of the RMC detector permitted scans to align the sample tube with the incident neutron beam.

Fig. 1. Two Samples, natural (top) and 2.7 % UO_2 (bottom), and a blank canister.

A high-speed beam shutter was automated to expose each sample to the beamline for a 60 *s* irradiation which was followed by the recording of delayed neutron emissions up to 180 *s* after the termination of irradiation. Total delayed neutron counts are recorded in 250 *ms* intervals.

The beam shutter can transition from fully open to closed in < 250 *ms* so only the first point in each run is discarded. This irradiation and counting process was repeated 50 times for the blank sample and up to 362 times for the sample containing 1.7 % enriched uranium, described in Table 1. The cumulative counts for each time channel were saved for analysis.

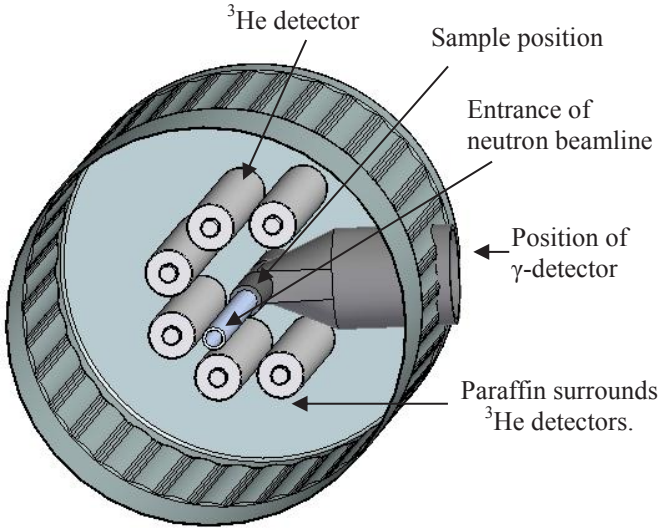

Fig. 2. Arrangement of detectors and sample in the NRU reactor beamline.

Table 1: ^{235}U content in samples and number of irradiation and counting cycles

Sample	^{235}U Mass	Cycles
2.7 % i	0.2676 *g*	251
2.7 % ii		242
1.7 % i	0.1557 *g*	362
		255
Nat U	0.0865 *g*	289
Blank	0.0000 *g*	50

DATA ANALYSIS

Temporal Behaviour

Of particular interest to nuclear forensics and safeguards is the capability to non-destructively identify and quantify fissile content. This can be accomplished via delayed neutron counting by solving the overdetermined linear system of equations below,

$$\begin{bmatrix} c_1 \\ \vdots \\ c_k \\ \vdots \\ c_n \end{bmatrix} = \begin{bmatrix} Q_{1_{233_U}} & Q_{1_{235_U}} & Q_{1_{239_{Pu}}} \\ \vdots & \vdots & \vdots \\ Q_{k_{233_U}} & Q_{k_{235_U}} & Q_{k_{239_{Pu}}} \\ \vdots & \vdots & \vdots \\ Q_{n_{233_U}} & Q_{n_{235_U}} & Q_{n_{239_{Pu}}} \end{bmatrix} \cdot \begin{bmatrix} m_{233_U} \\ m_{235_U} \\ m_{239_{Pu}} \end{bmatrix} \quad (1)$$

where c_k is the recorded delayed neutron count rate at time k, $[s^{-1}]$, m_j is the fissile mass of isotope j, $[g]$, and Q_j is the theoretical delayed neutron emission rate of one gram of a fissile isotope at time t $[s^{-1}g^{-1}]$, which can be described by Eq. (2) [5].

$$Q(t)_j = \frac{\varepsilon \upsilon_j N_A \sigma_{f_j} \Phi}{M_j} \sum_{i=1}^{8} \beta_{i_j} (1 - e^{-\lambda_i t_{irr}})(e^{-\lambda_i t}) \quad (2)$$

In this equation ε is the detection efficiency, υ_j is the total number of neutrons produced per fission, N_A is Avogadro's number $[mol^{-1}]$, σ_{f_j} is the thermal fission cross section $[cm^2]$, Φ the neutron flux $[cm^{-2}\ s^{-1}]$, M_j the molar mass $[g\ mol^{-1}]$, β_{i_j} the delayed neutron fraction of group i, t_{irr} the irradiation time, t the count time and λ_i the decay constant of group i $[s^{-1}]$.

Several systems have previously demonstrated the capability to distinguish between ^{235}U and ^{239}Pu content in aqueous solutions using delayed neutron counting [5,6], and this is the eventual goal of this project. However, in the work described here the only fissile isotope assumed to be present was ^{235}U. Therefore, equation 1 was greatly simplified to include only terms relevant to ^{235}U delayed neutron emission (highlighted in green in equation 1). The delayed-neutron eight-group parameters from the

International Atomic Energy Agency's Handbook of Nuclear Data for Safeguards [7] were used.

Fissile Magnitude

The beamline neutron flux magnitude at the sample's position is not well characterized, necessitating the requirement to use a fissile standard to determine absolute fissile mass in the samples analyzed. In the case of this work, the 251 cycle run examining the 2.7% enriched uranium sample was used for calibration. It was selected as it had the lowest uncertainties due to its increased fissile mass (0.2706 *g* of ^{235}U) and resulting high count rates.

RESULTS AND DISCUSSION

^{235}U Identification

Measurements were found to have a substantial neutron background of up to 636 ± 8 counts s^{-1}, which was subtracted from each recorded data set. The resulting delayed neutron count rates for each sample are shown in Figure 3. Also shown in Figure 3 is the linear least squares solution to Eq. (1) (assuming only ^{235}U content was present) for each sample.

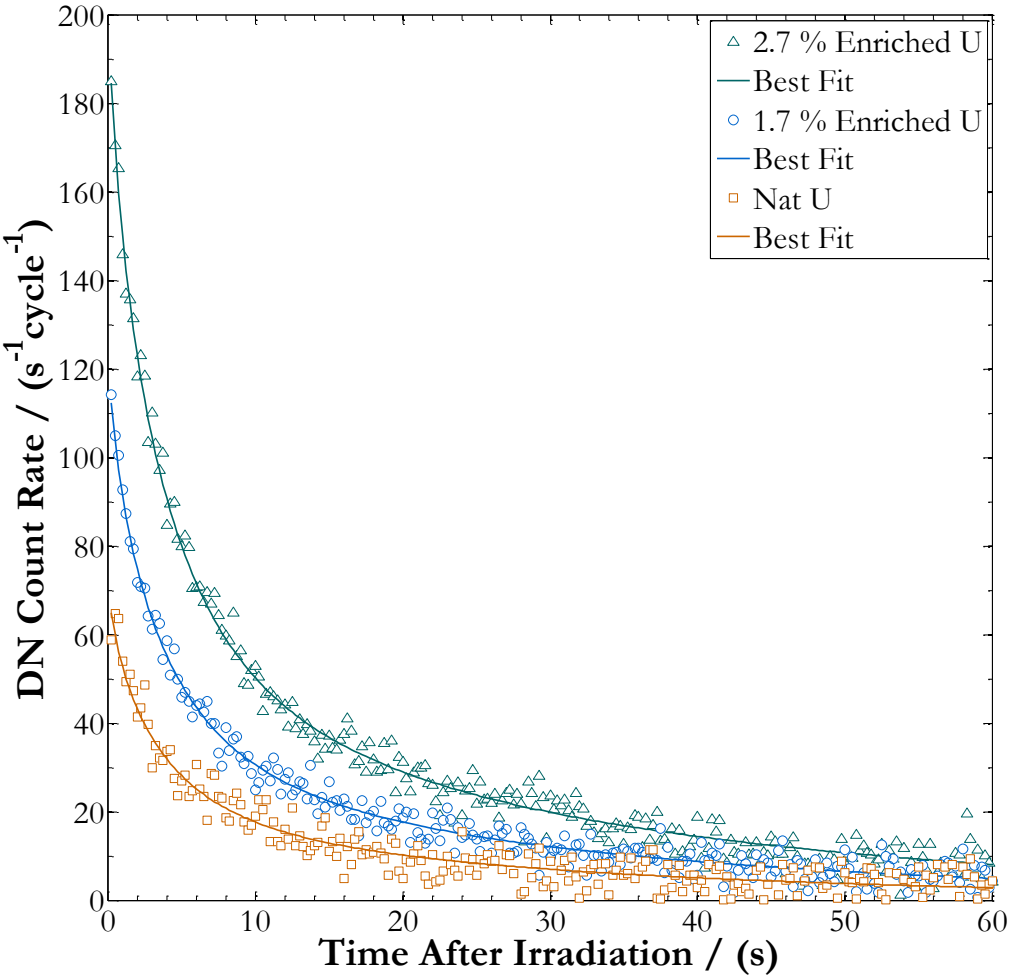

Fig. 3. Delayed Neutron (DN) count rate and linear least squares fit of Eqn 1 (for ^{235}U content only).

The total mass of ^{235}U was determined using a 2.7 % enriched run as a standard, the results are shown in Table 2. For this experiment the irradiation was controlled by time rather than the fluence, consequently variations in reactor power likely explains the deviation in relative error.

The detector configurations used in previous works [1, 2] was purpose-built for different applications and environments. One of the objectives of this exercise was to assess the two concepts in order to determine the best compromise of design requirements for special nuclear material identification. The primary deficit of the CNL [1] design was the very low solid angle that was a consequence of efforts to reduce ambient background and stray neutrons. The RMC [2] design subtends a much greater solid angle and incorporates more detectors. The orders of magnitude improvement in count rate clearly indicates that a marriage of the concepts can lead to a high-performing delayed neutron detector for in-beam irradiations.

Table 2: Actual and experimentally determined ^{235}U amount via delayed neutron counting at the NRU.

Sample	Actual ^{235}U (g)	Exp ^{235}U (g)	% Relative Error
2.7 % i	0.2676	-standard-	
2.7 % ii		0.2578	-3.7
1.7 % i	0.1557	0.1569	0.8
1.7 % ii		0.1578	1.4
Nat U	0.0865	0.0906	4.7

CONCLUSIONS & FUTURE WORK

In this work, in-beam delayed neutron experiments were used to determine ^{235}U mass with an average absolute error of ± 2.7 *%.* This error is lower than that obtained at RMCC for the assay of ^{235}U content in aqueous solutions (3.6 %) using delayed neutron counting. Although this analysis only includes the mass determination of one fissile isotope, the errors are in line with those of ^{235}U measurements in bulk samples by Myers *et al* [3] and environmental standards (soil and sediment) obtained by NIST using delayed neutron counting [8]. This instrumentation and analysis will be further challenged when experimentation is expanded to assay of mixed-oxide (^{235}U and ^{239}Pu) and uranium/thoria (^{233}U) fuel.

The strengths of the present arrangement include its ability to accommodate samples of greater and more irregular volume than those irradiated within the SLOWPOKE reactor at RMC, and the facile collection of cumulative decay data from repetitive irradiations. Whilst the lower flux in the beam for these measurements at CNL did not lend itself to trace analysis, it could prove beneficial in the analysis of large or highly enriched samples. Moreover, choice of beamline and neutron optics could lead to factor of 10 or 100 increase on the flux delivered to the sample. In-beam work is also more amenable to examination of samples of unknown origin or composition as they would not have to be sent adjacent to the core of a nuclear reactor.

Delayed neutron counting has been demonstrated to be a rapid, accurate, and precise method for special nuclear material detection and identification. RMCC aims to use this technique in upcoming nuclear forensic round robins to assess its possible contributions to Canadian safeguards efforts.

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