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POST-IRRADIATION NEUTRON EMISSIONS FROM CANDU FUELS AND THEIR NUCLEAR FORENSICS APPLICATIONS

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ABSTRACT – Fissile materials within a fuel pellet can be discerned rapidly and non-destructively via the analysis of post-irradiation delayed neutron emissions. The delayed neutron counting technique is well established within the Canadian nuclear industry, and uses include the detection of defective CANDU fuel. This work discusses these neutron emissions from CANDU fuel in the context of detection and attribution. Monte Carlo simulations of these emissions from current and proposed CANDU fuels (thoria and mix oxide based) have been performed. These simulations are compared to measurements of delayed neutron emissions from ^{233}U and ^{235}U, and the feasibility of CANDU fuel characterization is discussed.

1. Introduction

Nuclear Forensic Analysis (NFA) uses a variety of analytical techniques and instrumentation to characterize nuclear materials with the eventual goal of their attribution [1]. The assay of special nuclear materials (SNM), ^{233}U, ^{235}U and ^{239}Pu, has been particularly emphasized as these materials may be diverted for illicit means [2]. The Royal Military College of Canada (RMCC) is part of an international technical working group concerned with enhancing and evaluating the NFA capabilities of government laboratories in Canada and other member states [3]. The RMCC has the necessary licenses to work with SNM, in addition to varied nuclear instrumentation including: inductively coupled plasma mass spectroscopy, gamma and alpha spectroscopy, liquid scintillation counting and more recently, delayed neutron counting.

Delayed neutron counting is currently employed by Canadian industry to determine uranium content in geological samples and to detect failed fuel in CANDU reactors. A delayed neutron counting (DNC) system was developed and implemented at the RMCC with the specific intent of analyzing fissile content for NFA applications [4]. The DNC system has been validated for the analysis of samples containing just one known fissile isotope via the examination of delayed neutron (DN) temporal behaviour. This research is currently expanding to record the DNs emitted after the irradiation of binary fissile mixtures.

This paper uses previously published work on DN analysis at the RMCC to extend these efforts into CANDU fuel analysis applications. If diverted CANDU fuel were to be intercepted by authorities, the prompt and non-destructive characterization of the fissile content could assist in determining a source of origin. Such information would be of valuable in law enforcement and security contexts. Building on work presented at the 2012 Canadian Nuclear Society Student Conference in Saskatoon, SK, the behaviour of DN signatures produced by various CANDU fuels is modelled in MCNP6 and compared to experimental measurements conducted at the RMCC.

2. Theory

If a fuel pellet were irradiated in the SLOWPOKE-2 reactor at the RMCC, the (*n,f*) interaction with the fissile materials present would result in the release of prompt neutrons and fission products. Many of these fission products are denoted as DN precursors as they will undergo β^- decay and produce a daughter isotope with an unbound neutron. This results in the release of delayed neutrons at a rate consistent with the half-life of their associated precursor. DNs are often grouped according to their half-lives [5] and given an associated yield, α_i, which is characteristic of the isotope, *j*, undergoing fission. The total count rate, *S(t)* can be approximated as shown in Eqn. 1 [6]:

$$S(t) = \sum_{j=1}^{n}[\frac{\varepsilon\sigma_{f_j}\nu_j\Phi N_A}{M_j}\sum_{i=1}^{8}\alpha_{ij}(1-e^{-\lambda_i t_{irr}})(e^{-\lambda_i t_d})(e^{-\lambda_i t})]m_j + B(t) \quad (1)$$

Where *n* is the number of fissile isotopes present in a sample, m_j is the mass of the fissile isotope *j* [g], ε is efficiency of neutron detection, σ_f is the fission cross section [b], ν_j is the number of DNs produced in the fission of that isotope, Φ is the neutron flux [$cm^{-2}s^{-1}$], N_A is Avogadro's number [mol^{-1}], M_j the isotope's molecular mass [g mol^{-1}], λ_i is the decay constant for group *i* [s^{-1}], t_{irr} the irradiation duration of the sample [s], t_d the decay time of the sample [s], *t* the count time [s], and *B(t)* is the neutron background of the system.

Therefore in the case of a fully characterized system (where flux, efficiency and neutron background are well established), the only unknown in Eq. 1 is the masses of fissile isotopes present. In the case of a sample containing two fissile isotopes, i.e. ^{233}U and ^{235}U, the count rate can be written as:

$$S(t) = [Q(t)_{233_U}\ Q(t)_{235_U}] \cdot \begin{bmatrix} m_{233_U} \\ m_{235_U} \end{bmatrix} + B(t) \quad (2)$$

Where

$$Q(t)_j = [\frac{\varepsilon\sigma_{f_j}\nu_j\Phi N_A}{M_j}\sum_{i=1}^{8}\alpha_{ij}(1-e^{-\lambda_i t_{irr}})(e^{-\lambda_i t_d})(e^{-\lambda_i t})] \quad (3)$$

Thus, the relative quantities and total fissile content in a specimen can be determined via a least squares fit to experimental measurements.

3. Experimental

Samples containing fissile content were prepared from natural uranium (CRM 4321C, NIST, Gaithersburg, MD), and 99 atom % ^{233}U (CRM 111-A, 99.49 atom% ^{233}U, New Brunswick Laboratory, Argonne, IL). The fissile content was dissolved in HNO_3 (Optima, ThermoFisher Scientific, Ottawa, ON) and further diluted by distilled deionised water. Each sample contained various combinations of ^{233}U and ^{235}U (each fissile solution was contained its own 1.5 *ml* polyethylene vial). The individual aqueous fissile solutions were then heat sealed in a larger 7 *ml* polyethylene vial to create a *fissile mixture.*

After preparation, a sample was sent to the RMCC's SLOWPOKE-2 [7] reactor for a 60 *s* irradiation. Upon the expiration of this irradiation period, the sample was transferred via pneumatic tubing to a counting arrangement consisting of six ^{3}He detectors embedded in paraffin. The sample emits DNs isotropically, which are recorded by customized LabVIEW^{TM} software. The sample is then sent to a disposal unit where they can be retrieved for additional tests or safely disposed of at the end of the counting period. Further information on the experimental set up can be found in Reference 4.

4. The MCNP6 Model

MCNP6 [8] is the newest release of a Monte Carlo code developed by Los Alamos National Laboratory. The counting system described in the experimental section was modelled in this software and is shown in Figure 1. The irradiation and counting portions of experiments were reproduced using the fixed source option in MCNP6. This MCNP6 model was slightly modified from the original described in Reference 9 to accommodate a small piece of CANDU fuel of varying composition.

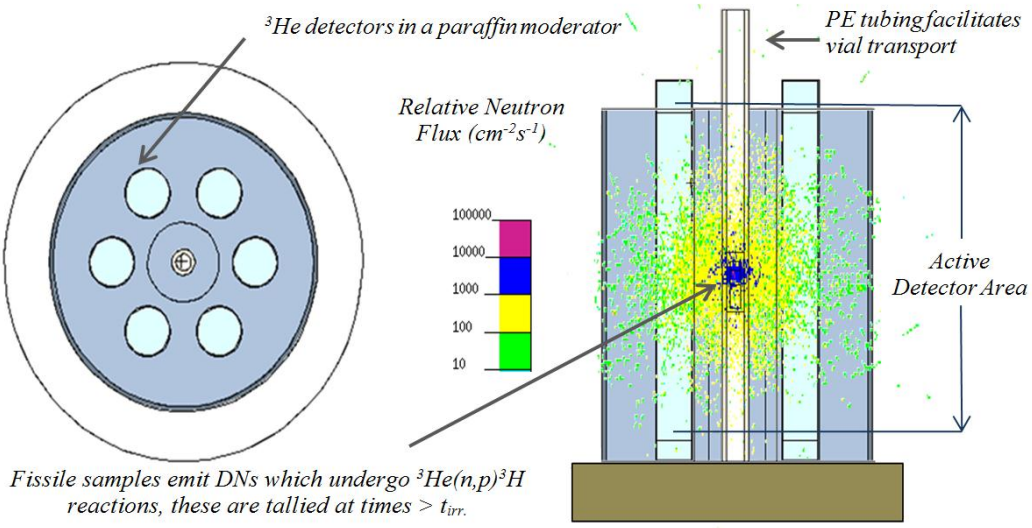

Figure 1 A Schematic of the Delayed Neutron Counting System [9]

5. Results and Discussion

5.1 Temporal Behaviour of ^{233}U and ^{235}U Delayed Neutrons: Simulations & Experiments

The assay of fissile mixtures via delayed neutron counting is dependent on the temporal behaviour of the emitted neutrons with count time. Dead time effects and timing uncertainties must be properly accounted for to reduce their distortion of the temporal behaviour of recorded DNs. Figure 2 depicts the measured and MCNP6 simulated temporal behaviour of small quantities of ^{233}U and ^{235}U. Figure 3 shows the same data, however differences in temporal behaviour are emphasized by normalizing all initial count rates to the same value. This normalization also eliminates uncertainties arising from flux, system efficiency characterizations and the fissile concentrations of solutions. Figure 3 shows a more rapid decay for ^{235}U (as it has relatively fewer long lived delayed neutron precursors) for both simulations and measurements.

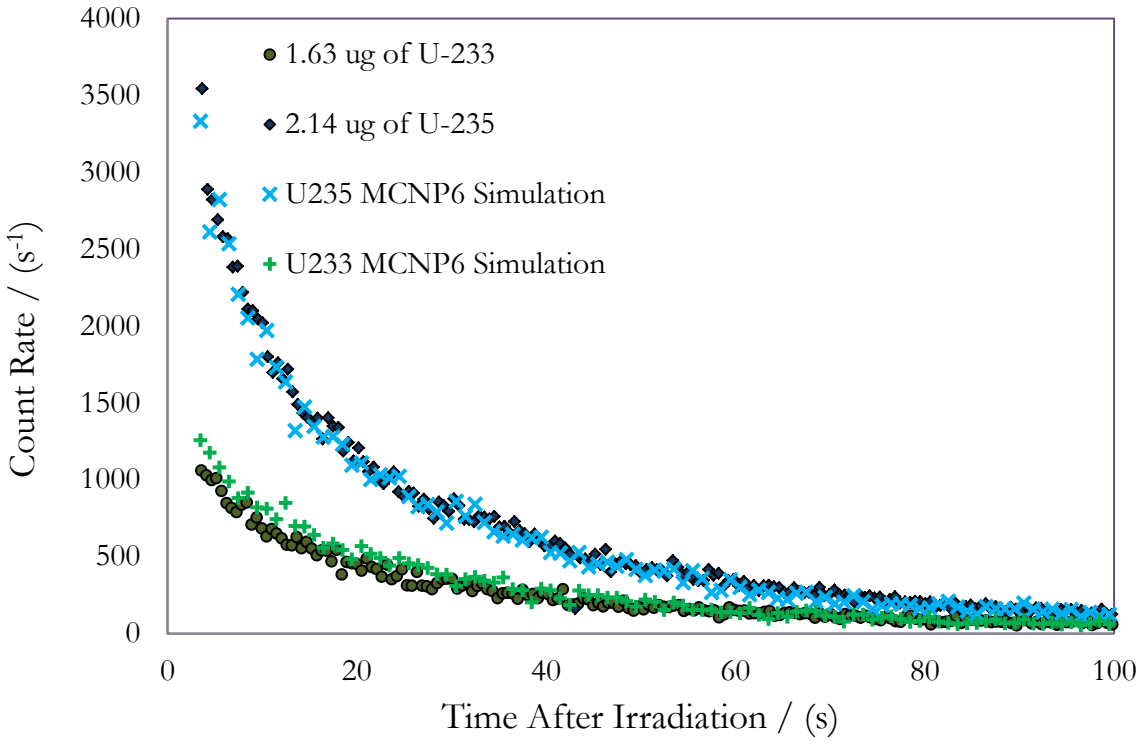

Figure 2: MCNP6 Delayed Neutron Behaviour & Experimental Measurements

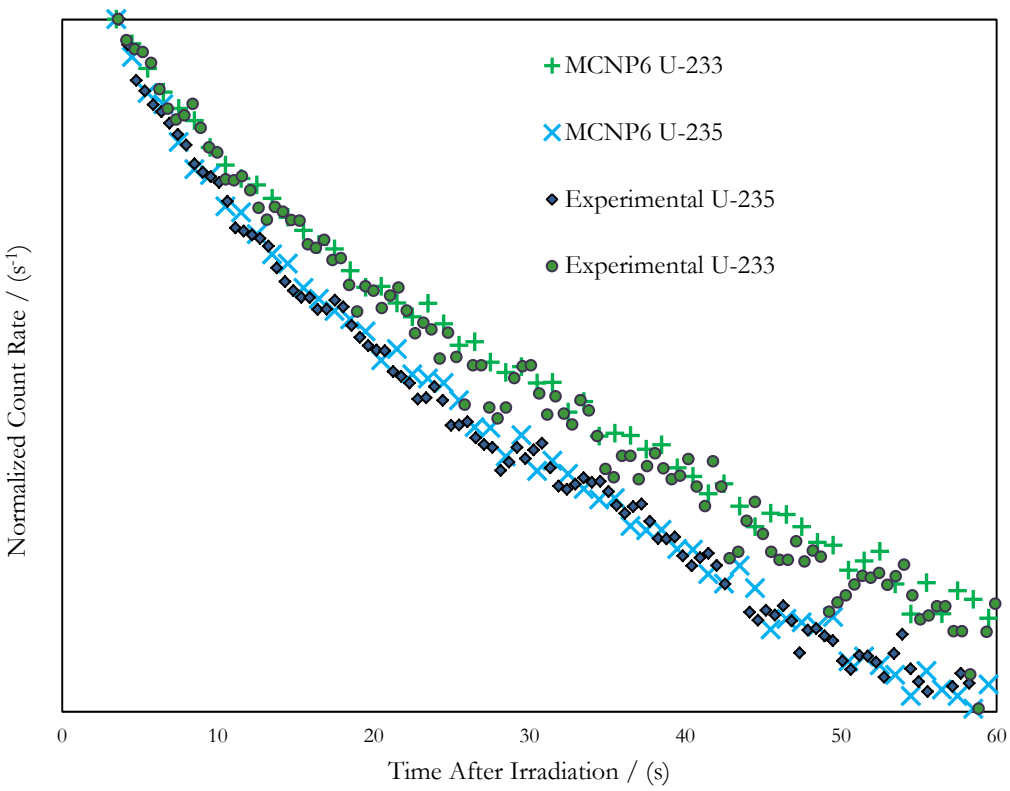

Figure 3: Normalized MCNP6 Delayed Neutron Behaviour & Experimental Measurements

5.2 Fissile Mixture Characterization via Delayed Neutron Counting

MCNP6 model predictions and measurements have confirmed the distinct DN temporal behaviour of ^{233}U and ^{235}U after their irradiation. Recent work [10] has attempted to quantify the relative ratios of ^{233}U and ^{235}U in mixtures. Table 1 shows the relative ratios of ^{233}U to ^{235}U as a percentage of total fissile mass, ranging from 0 % ^{233}U in Sample 1 to 100 % ^{233}U in Sample 10. The Experimental Ratio column shows the average results after solving Eq. (2) for $\begin{bmatrix} m_{233_U} \\ m_{235_U} \end{bmatrix}$. Each individual measurement was found to have a high degree of uncertainty, and the relative amounts of ^{233}U and ^{235}U had to be determined through duplicate measurements of each sample. This is easily facilitated due to the non-destructive and rapid nature of delayed neutron counting. The system was able to determine the ratio of ^{233}U content to that of ^{235}U after duplicate measurements, with an average absolute error of ± 4 %.

Table 1: DNC Determination of the Ratio of ^{233}U to ^{235}U in Fissile Mixtures [10]

Sample	Actual Ratio		Experimental Ratio		Sample	Actual Ratio		Experimental Ratio	
1	0 : 100		4 : 96	(± 3)	**6**	37 : 63	(± 1)	45 : 55	(± 8)
2	13.1 : 86.9	(± 0.3)	11 : 89	(± 4)	**7**	51 : 49	(± 2)	50 : 50	(± 6)
3	17.7 : 82.3	(± 0.3)	19 : 81	(± 5)	**8**	71 : 29	(± 1)	78 : 22	(± 6)
4	31.3 : 68.7	(± 0.6)	27 : 73	(± 4)	**9**	96 : 4	(± 4)	92 : 8	(± 2)
5	36 : 64	(± 1)	44 : 56	(± 4)	**10**	100 : 0		97 : 3	(± 2)

5.3 CANDU Fuel Applications

The MCNP6 model developed for previous work has been extended to simulate the investigation of three CANDU fuel pellet compositions in the DNC system. The first composition was natural UO_2 used in operational CANDU reactors. The second pellet fragment contained an oxide composition corresponding to a proposed thorium cycle [11]; it had both thorium and uranium content with the uranium isotopics consisting of 62 % ^{233}U, 23 % ^{234}U, 6 % ^{235}U, and 9 % ^{236}U. The third composition is a proposed mixed oxide (MOX) fuel containing 94 wt% ThO_2 and 6 wt% PuO_2 (with a ^{239}Pu to ^{240}Pu ratio of 15.7:1) [12]. Only a small fragment of each fuel pellet type was modelled, as this reflects the limitation with regard to the fissile content of a SLOWPOKE-2 irradiation site. The simulation assumed fresh, unirradiated fuel, thus any changes in fissile content as a function of fuel burnup were neglected. MCNP6 predicted significant temporal differences in the three types of fuel, they are shown in Figure 4.

The Monte Carlo models of the fuel pellets account for sub-critical multiplicity effects and matrix self-shielding, and results in unique temporal behaviours for each pellet of interest. Figure 5 shows a comparison of the predicted temporal decay behaviour of the natural UO_2 pellet and measurements of irradiated natural U solutions. A comparison of the normalized count rates shows excellent agreement between simulations and measurements. MCNP6 simulations of the Th/U oxide pellet and the accompanying ^{233}U measurements are also shown in Figure 5. However measurements only agree with simulations until ~count times of 60 *s*. The simulated pellet fragment contained 61 % ^{233}U and 6 % ^{235}U content whereas the irradiated sample had no ^{235}U content; this difference in ^{235}U content accounts for the discrepancy. Other fissionable isotopes were present in the simulations however these would produce negligible DNs when irradiated in the SLOWPOKE-2 due to the high thermal to fast flux ratio of the reactor.

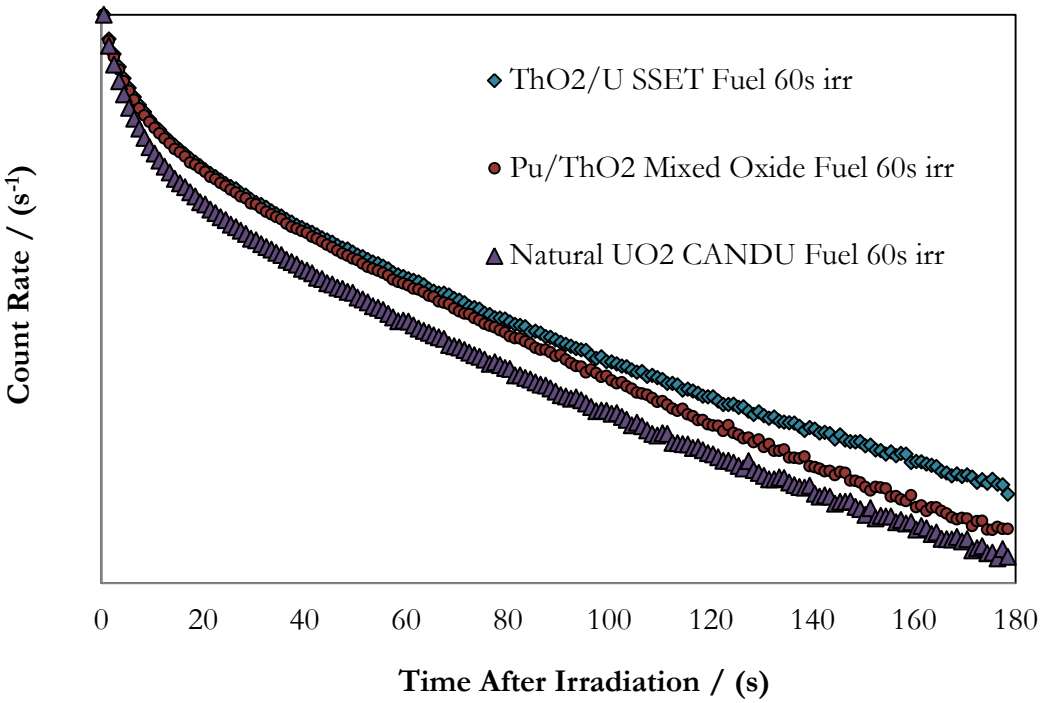

Figure 4: Simulated Temporal Behaviour of Present & Proposed CANDU Fuel Pellets

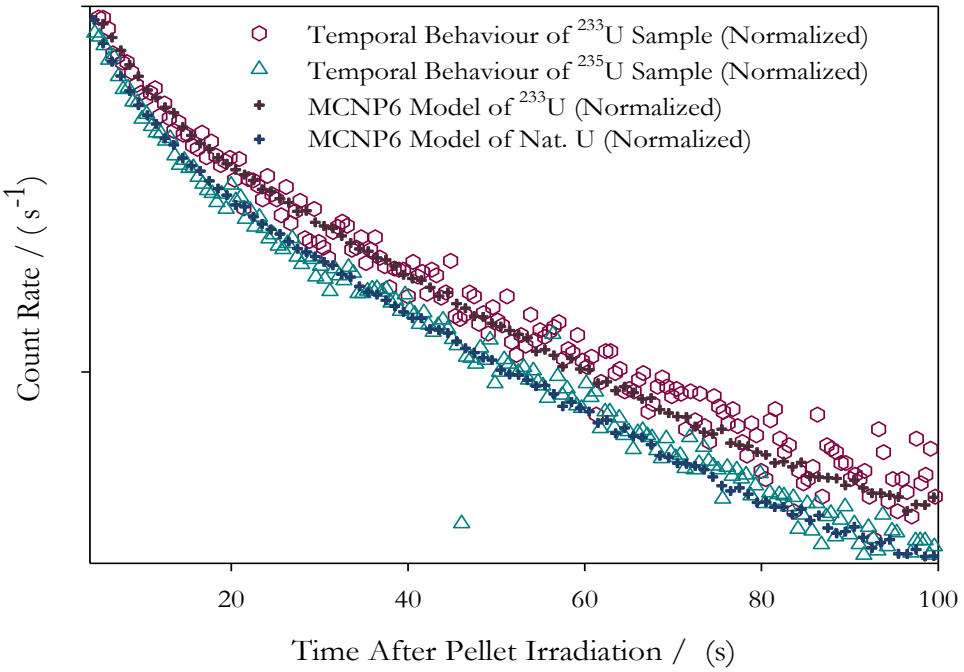

Figure 5: Fuel Pellet Simulations Compared to DNC system Measurements

6. Conclusions

Analysis of ^{233}U:^{235}U by DNC using the SLOWPOKE-2 Facility demonstrates that the collection and analysis of temporal data facilitates the determination of ^{233}U and ^{235}U fissile content in mixed systems with accuracy and precision that are acceptable for NFA. These data also demonstrate that a small mass of fissile content is required to perform DNC analysis. This supports the hypothesis that only a small portion of the fuel pellet would be required for analysis DNC, liberating the vast majority of the pellet for immediate NFA with other instrumentation. Moreover, the non-destructive nature of DNC allows for the preservation of the fragment as evidence in the case prosecution is required. Monte Carlo simulations provide a valuable modeling and development tool. These simulations have indicated the fuel pellets would produce DNs with comparable temporal behaviour to that measured at the RMCC. Although measurements conducted at the RMCC using unirradiated fuel have a far higher signal to noise background than would be expected in the analyses of irradiated fuel pellet fragments, instrument development is underway to limit the affects gamma-ray interference.

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