CANDU Fuel Attribution Through the Analysis of Delayed Neutron Temporal Behaviour

M.T. Sellers, E.C. Corcoran, D.G. Kelly*
Royal Military College of Canada, Kingston, Ontario, Canada, *David.Kelly@rmc.ca

Summary

Delayed Neutron Counting (DNC) is an established technique in the Canadian nuclear industry as it is used for the detection of defective fuel in several CANDU reactors and the assay of uranium in geological samples. This paper describes the possible expansion of DNC to the discipline of nuclear forensics analysis. The temporal behaviour of experimentally measured delayed neutron spectra were used to determine the relative contributions of ^{233}U and ^{235}U to the overall fissile content present in mixtures with average absolute errors of ±4 %. The characterization of fissile content in current and proposed CANDU fuels (natural UO_2, thoria and mixed oxide (MOX) based) by DNC analysis is evaluated through Monte Carlo simulations.

1. Introduction

Nuclear forensic analysis (NFA) is a modern science that employs many analytical techniques for the assay, characterization and attribution of nuclear materials [1]. The radionuclide debris of a nuclear explosion, the products of a radiological dispersal device detonation and nuclear materials intercepted from smuggling activities, all represent potential subjects of NFA. Simulated exercises have emphasized the requirement for the Canadian government to expand current nuclear forensics expertise, protocols and capabilities [2]. Emphasis has been placed on the analysis and attribution of special nuclear materials (SNM), which include the fissile isotopes ^{233}U, ^{235}U and ^{239}Pu. Delayed neutron counting (DNC) is an established technique capable of determining fissile isotope presence and quantity in a wide variety of environmental and synthetic matrices. The Canadian nuclear industry currently employs variants of this technique both in the determination of uranium content in geological samples and for the detection of failed fuel in several CANDU reactors.

The Royal Military College of Canada (RMCC) has developed a DNC system, which uses the SLOWPOKE-2 reactor to analyse samples containing fissile isotopes. Whereas more traditional DNC systems record the cumulative delayed neutron (DN) counts emitted during a designated pre-set time (commonly about one minute in duration), this system has been designed to record the temporal behaviour of the DNs produced after the irradiation of samples. By recording the temporal behaviour of the DNs, the system aims to distinguish between those produced from the fission of ^{233}U, ^{235}U, and ^{239}Pu. The system has been established for the analysis of ^{235}U [3]. Current assessments focus on matrices containing ^{233}U, ^{239}Pu, and binary and tertiary fissile mixtures. As DNC analysis is rapid, non-destructive and capable of assaying fissile content in a variety of matrices, this complementary technique has been integrated with existing analytical instrumentation at RMCC for the analysis of nuclear materials. This paper explores the current capability of the DNC system at RMCC to distinguish between the DNs produced from ^{233}U and those of ^{235}U, through the assessment of their temporal behaviour.

The relevance to the Canadian nuclear industry is illustrated by a comparison of three CANDU fuel pellets. The first example contains natural UO_2 as used in current reactor designs. A second has an oxide composition corresponding to a proposed thorium cycle. This pellet contains both thorium and uranium content, with the uranium isotopic composition consisting of 62 % ^{233}U, 23 % ^{234}U, 6 % ^{235}U,

and 9 % ^{236}U [4]. The final example is a proposed mixed oxide (MOX) fuel containing 94 wt% ThO_2 and 6 wt% PuO_2 (with the ratio of ^{239}Pu to ^{240}Pu of 15.7:1) [5]. The interception of illicitly trafficked material would require prompt identification. The analysis of this material through non-destructive means, for example, DNC, may help determine the origin and identity of such material. The expected behaviour of DNs produced by fuel pellets, if analysed at RMCC, is modelled in the simulation software MCNP6 [6], and the temporal behaviour of these pellets is compared to experimental DNC system measurements.

2. Delayed Neutron Counting Theory

The neutron-fission (*n,f*) interaction of fissile isotope *j* results in the immediate release of prompt neutrons and fission fragments. Delayed neutrons are produced immediately after the β^- decay of some fission products (denoted as delayed neutron precursors) with half-lives dictated by the β^- process. The many delayed neutron precursors are grouped by half-lives and relative yields, α_i. The magnitude of DNs produced and their temporal behaviour is unique to each fissile isotope, due to their different fission cross sections and fission product yields. Therefore, the temporal behaviour of the DNs produced will be a function of fissile isotopes present and their relative quantities. The DN count rate, *S(t)*, after the irradiation of *n* fissile isotopes is a superposition of each isotope's DN production and the system background *B(t)* [7] :

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

Where in Eq. (1) m_j is the mass of the fissile isotope *j* [g], ε is efficiency of the instrumentation, σ_f is the thermal fission cross section [b], ν_j is the number of DNs produced in the fission of that isotope, Φ is the thermal 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 before the commencement of counting [s], and *t* the count time [s]. Therefore in the analysis of fissile mixtures in a fully characterized system, the only unknowns in Eq. (1) are the masses of the fissile isotopes present, m_j. If the DNs produced by a sample are recorded as a function of time, the individual fissile masses present may be discerned through the application of Eq. (1) to the experimental data.

3. Experimental

Ref. [3] contains a detailed description of the DNC system at RMCC. Each sample was irradiated in the SLOWPOKE-2 Facility reactor for 60 s in a predominately thermal neutron flux. After irradiation the samples were sent via pneumatic tubing to an array of ^3He detectors, which recorded the DN count rate in 0.5 s intervals. The DN signal was recorded for 180 s and was subsequently imported into fissile isotope analysis software written in Matlab™ (R2011a, Mathworks, Natick, MA). This software corrects for the neutron background, and electrical dead time effects. The count rate recorded by the apparatus was compared to the expected behaviour of these fissile isotopes to determine the actual amount of ^{233}U and ^{235}U in each sample. The samples 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) that were dissolved in HNO_3 (Optima, ThermoFisher Scientific, Ottawa, ON). The total fissile content in each sample was a combination of both the ^{233}U and ^{235}U solutions.

4. Results and Discussion

4.1 A Comparison of the Temporal Behaviour of ^{233}U and ^{235}U Delayed Neutrons

An MCNP6 model of the DNC system and procedure outlined in Section 3 has been developed in collaboration with Los Alamos National Laboratory and is described in Ref. [8]. Figure 1 shows the simulated and measured DN temporal behaviour for aqueous samples containing ^{233}U and ^{235}U. Differences in the magnitude of the MCNP6 and experimental results can be attributed to experimental uncertainties that include; solution fissile concentrations, delay timing and system efficiency uncertainties. The upper right corner of Figure 1 shows the normalized spectra, which emphasize the differences in the temporal behaviour of ^{233}U and ^{235}U after the count rates for each isotope have been normalized to an identical starting rate. As evident in Figure 1, ^{235}U DN count rate decays more rapidly than ^{233}U; an observation that is consistent with the smaller fraction of longer lived DN precursors in ^{235}U.

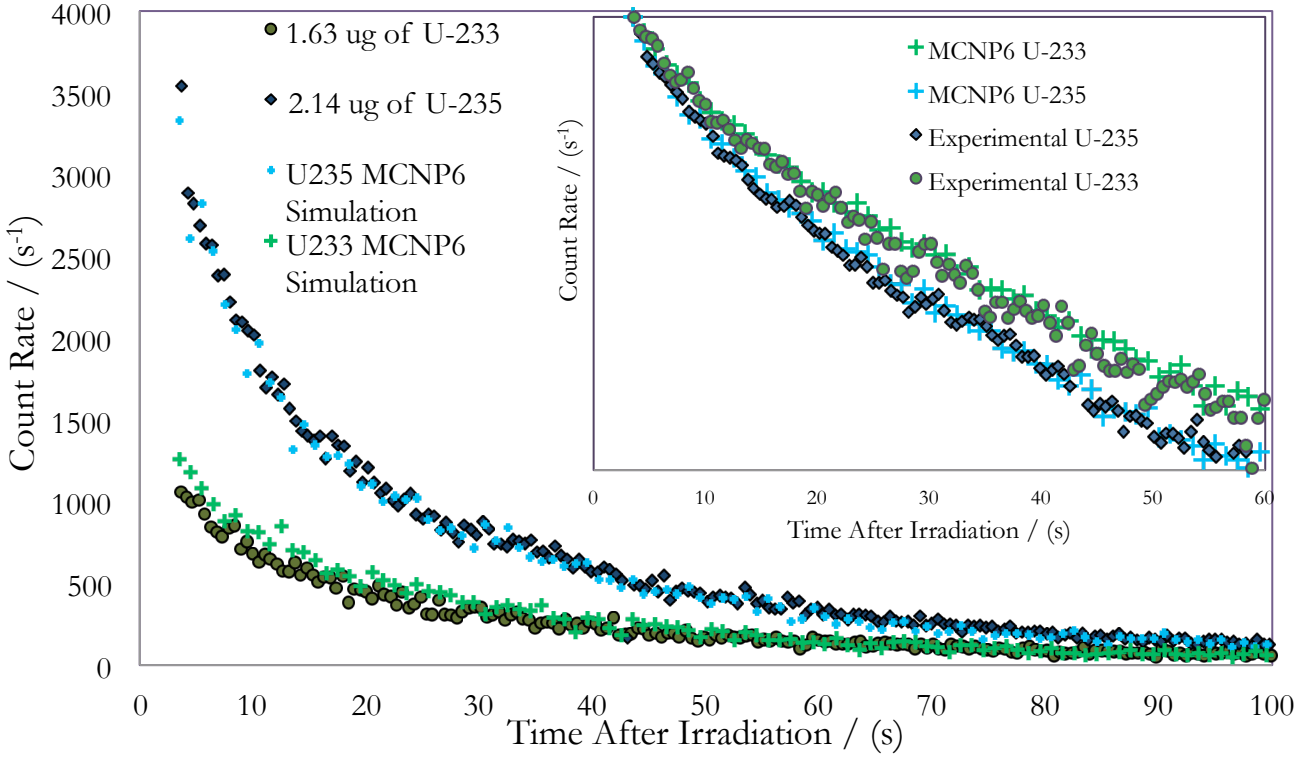

Figure 1: MCNP6 Delayed Neutron Behaviour & Experimental Measurements.

4.2 The Differentiation of ^{233}U and ^{235}U Fissile Content through Delayed Neutron Counting

Experimental measurements and MCNP6 model predictions have established a distinct DN temporal behaviour of fissile mixtures dependent on the relative composition of ^{233}U and ^{235}U. This temporal behaviour was assessed to quantify the relative ratios of the fissile isotopes present in 10 samples containing a combination of ^{233}U and ^{235}U. Table 1 shows the actual ratio of ^{233}U to ^{235}U in each sample and the experimental results obtained through DNC analysis. It was found that an individual measurement of each sample had a high degree of uncertainty and the relative amounts of ^{233}U and ^{235}U

were therefore determined through duplicate measurements of each sample. The DNC system was able to determine the relative ratio of ^{233}U content to that of ^{235}U present in each sample with a mean absolute error of ±4 %.

Table 1: DNC Determination of the Ratio of ^{233}U to ^{235}U in Fissile Mixtures [9]

Sample	Actual Ratio		Experimental Ratio	
1	0 : 100		4 : 96	(± 3)
2	13.1 : 86.9	(± 0.3)	11 : 89	(± 4)
3	17.7 : 82.3	(± 0.3)	19 : 81	(± 5)
4	31.3 : 68.7	(± 0.6)	27 : 73	(± 4)
5	36 : 64	(± 1)	44 : 56	(± 4)

Sample	Actual Ratio		Experimental Ratio	
6	37 : 63	(± 1)	45 : 55	(± 8)
7	51 : 49	(± 2)	50 : 50	(± 6)
8	71 : 29	(± 1)	78 : 22	(± 6)
9	96 : 4	(± 4)	92 : 8	(± 2)
10	100 : 0		97 : 3	(± 2)

4.3 Applications to Current & Proposed CANDU Fuel Attribution

The MCNP6 model of the DNC system was also used to simulate the investigation of three CANDU pellet compositions in the DNC system at RMCC as shown in Figure 2. A small fragment of each fuel pellet type (nat. UO_2, ThO_2/U fuel and Pu/ThO_2 MOX fuel) replaced the aqueous solutions in the previous MCNP6 models. It was assumed in the MCNP6 model that the fuel fragment analyzed was from fresh, un-irradiated fuel, allowing changes in fissile content as a function of fuel burnup to be neglected in this model. Due to the differences in the fissile content of each fuel, the temporal behaviour was quite distinct. Also depicted in Figure 2, for comparison purposes, is the measured behaviour of samples containing only ^{233}U or ^{235}U. Note the experimental results have been scaled to emphasize differences in temporal behaviour rather than the magnitude of DNs produced.

The MCNP6 models of the CANDU fuels account for matrix self-shielding and subcritical multiplicity effects yet still display distinct temporal behaviours resultant from the varying fissile content. A comparison of the experimental temporal behaviour of the measurements of natural U samples at RMCC with that predicted by the irradiation of the natural UO_2 fuel fragment shows excellent consistency. The distinct behaviour of the Th/U oxide pellet model relative to either ^{233}U or natural uranium is apparent, Figure 2. The differences between this fuel and a natural UO_2 pellet are readily observed (main display), whilst the deviations from the ^{233}U sample at count times are clear at > 60 s (insert). The respective ^{233}U and ^{235}U contents of 61 % and 6 % fully justify these data. Furthermore, since ^{235}U displays a lower relative DN contribution at > 60 s compared with ^{233}U, this would result in the Th/U pellet temporal behaviour decaying at a slightly greater pace than experimental measurements of a pure ^{233}U sample, as seen in Figure 2 insert.

While samples containing the exact isotopic compositions described for current and proposed CANDU fuel pellets were not experimentally measured, evaluations of the ability of the DNC to distinguish fissile mixtures can be made using the data presented in Table 1. The DNC system was able to predict the fissile distribution of ^{233}U to that of ^{235}U with a mean absolute error of ±4 %. The relatively small amounts of fissile samples measured (*ca.* 1-2 μg) indicate only a small portion of the fuel pellet would be required for analysis through DNC. This fragment could also be preserved as evidence because of the non-destructive nature of the instrumentation. MCNP6 models indicate CANDU fuel pellets would produce DNs with temporal behaviour comparable to that observed in measurements performed at RMCC. Experiments showed that the analyses of each sample should be performed in duplicate or

triplicate measurements to reduce the uncertainty when determining fissile distributions. Such a protocol is simple because of the rapid and non-destructive nature of the technique, which facilitate re-analysis of the same material. Extrapolation of the data presented in Table 1 indicate the system is capable of detecting the presence of multiple fissile isotopes; a capability which would be ideal when analysing a fuel pellet with unknown origin.

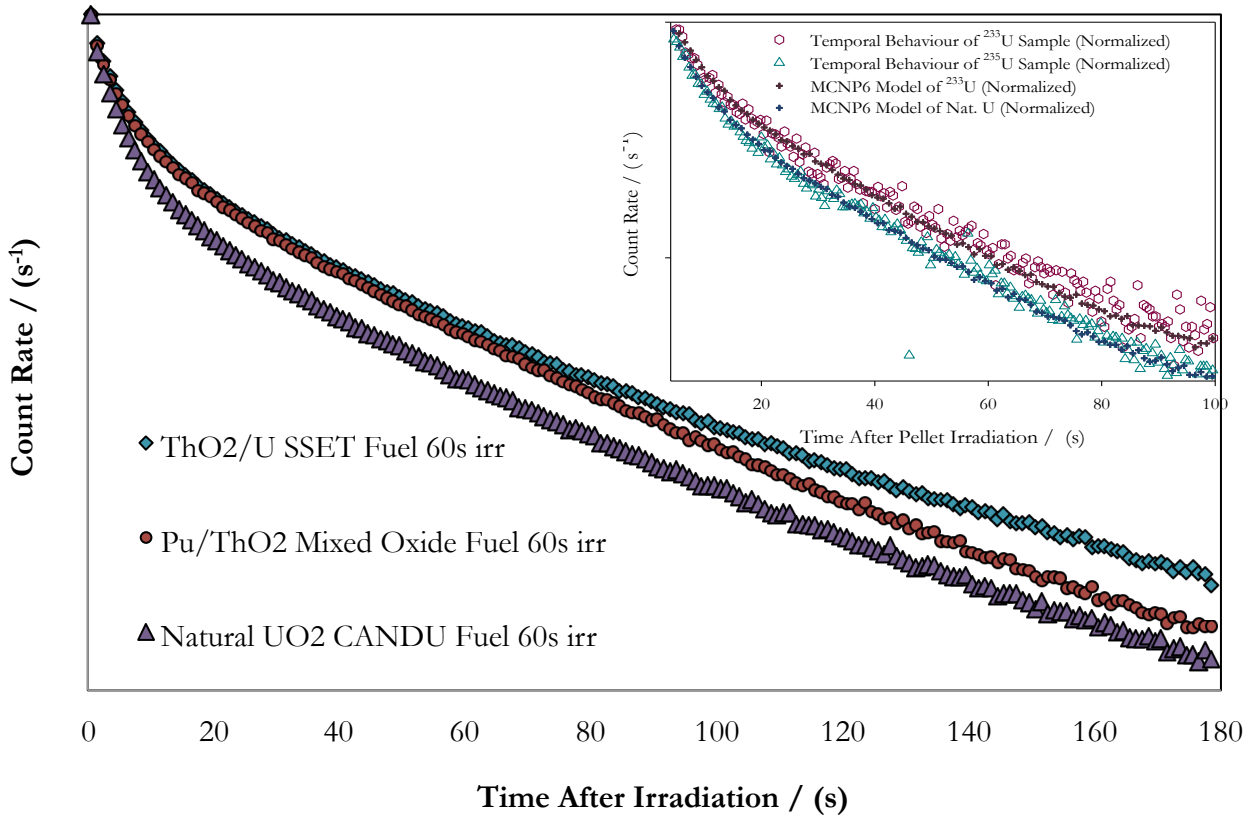

Figure 2: A Simulated Temporal Behaviour of Present & Proposed CANDU Fuel Pellets & A Comparison to DNC system Measurements (inset)

5. Future Work & Conclusions

Experimentation using ^{239}Pu contained in aqueous solutions is currently underway and will be expanded to include the analysis of $^{239}Pu/^{235}U$ and $^{239}Pu/^{233}U$ mixtures. ^{239}Pu measurements will be compared to the Pu/ThO_2 MOX fuel model results presented in Figure 2. Further work on the refinement of the technique could establish its feasibility as a form of fuel analysis; the determination of relative fissile compositions could identify the type of fuel material or its burnup.

Initial steps to determine the fissile content in mixtures of ^{233}U and ^{235}U have been undertaken at RMCC. The DNC system has been used to analyse the temporal behaviour of the DNs to determine the ratio of the ^{233}U to that of the total fissile mass present. A protocol using duplicate sample analyses resulted in a mean absolute error of ±4 %. The system at RMCC will undergo further refinement in an attempt to increase its accuracy and reduce associated uncertainties. The development of this technique will complement existing analytical instrumentation at RMCC, being non-destructive and capable of detecting the presence of SNM in NFA applications.

6. Acknowledgements

The authors wish to thank the Director General of Nuclear Safety, NSERC, and LANL for the financial support of this project. Also, the assistance of H.W. Bonin, T. Goorley, S. White, K. Nielsen, and D. Ferguson is appreciated.

7. References

[1] International Atomic Energy Agency "Nuclear Forensics Support" Vienna, 2006, IAEA Nuclear Security Series No. 2. Technical Guidance.
[2] C.L. Larsson, D.S. Haslip, "Consolidated Canadian results to the HEU Round Robin Exercise", *Defence R & D Canada Technical Memorandum,* 2004, TM 2004-192.
[3] M.T. Sellers, D.G. Kelly, and E.C. Corcoran, "An Automated Delayed Neutron Counting System for Mass Determinations of Special Nuclear Materials", *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 291 Iss. 2, 2012 pp. 281 – 285.
[4] H.W. Bonin, "CANDU Pressurized Heavy Water Reactor Throium-^{233}U Oxide Fuel Evaluation Based on Optimal Fuel Management" *Journal of Nuclear Technology* Vol. 76, 1987, pp. 390-399.
[5] S. Sahin, K. Yildiz, H. Sahin, N. Sahin, A. Acir, "Increased fuel burn up in a CANDU thorium reactor using weapon grade plutonium" *Journal of Nuclear Engineering and Design,* Vol. 236, Iss. 17, 2006, pp. 1778-1788.
[6] T. Goorley, M. James, T. Booth, F. Brown, J. Bull, L.J. Cox, J. Durkee, J. Elson, M. Fensin, R.A. Forster, J. Hendricks, H.G. Hughes, R. Johns, B. Kiedrowski, R. Martz, S. Mashnik, G. McKinney, D. Pelowitz, R. Prael, J. Sweezy, L. Waters, T. Wilcox, T. Zukaitis, "Initial MCNP6 Release Overview" *Submitted to the Journal of Nuclear Technology*, December 2011.
[7] X. Li, R. Henkelmann, and R. Baumgärtner, "Rapid Determination of uranium and plutonium content in mixtures through measurement of intensity-time curve of delayed neutrons," *Journal of Nuclear Instruments and Methods in Physics Research B*, Vol. 215, 2004, pp. 246 -251.
[8] M.T. Sellers, J.T Goorley, D.G. Kelly, E.C. Corcoran, "A Preliminary Comparison of MCNP6 Delayed Neutron Emission from ^{235}U and Experimental Measurements", Submitted to ANS 2012, Chicago, Illinois, USA, 2012 June 24 – 28.
[9] M.T. Sellers, E.C Corcoran, D.G. Kelly, "Simultaneous ^{233}U and ^{235}U Characterization Through the Assay of Delayed Neutron Temporal Behaviour", Accepted paper at PHYSOR 2012, Knoxville, Tennessee, USA, 2012 April 15-20.