An Automated Delayed Neutron Counting System for Mass Determination of Fissile Isotopes in Special Nuclear Materials at the Royal Military College of Canada

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Abstract

A Delayed Neutron Counting System (DNCS) has been designed and installed to enhance the ability of Royal Military College of Canada (RMC) to analyze Special Nuclear Materials (SNM). By detecting and recording delayed neutron counts from SNM samples irradiated by the SLOWPOKE-2 Reactor Facility at RMC, the DNCS is able to determine the masses of multiple fissile nuclei in the samples efficiently and accurately. The system is controlled by LabVIEW software. A separate mathematical program has also been developed to determine the mass of fissile nuclei present in SNM. The DNCS and fissile mass determination program have been validated using powdered UO_2 samples.

1. Introduction

Recent reports [1, 2] have highlighted the need for nations to establish nuclear forensic protocols and develop an efficient analysis procedure to aid in first response, post-event recovery, criminal prosecution and other law enforcement requirements. The Royal Military College of Canada (RMC) is currently enhancing its nuclear forensic capabilities in addition to developing protocol for the analysis of Special Nuclear Materials (SNM) using the resources of the SLOWPOKE-2 Reactor Facility at RMC. To enhance current capabilities at the Facility, a Delayed Neutron Counting System (DNCS) was designed and installed in the summer and fall of 2009. This technique utilizes the SLOWPOKE-2 Reactor Facility at RMC to provide a rapid and non-destructive capability to determine accurately the masses of the fissile isotopes present in Special Nuclear Material samples. This system also meets current commercial needs, as it will be able to provide elemental analysis for uranium, even in complex matrices, in a matter of minutes rather than the several days currently required for gamma-ray spectrometric analysis. To achieve a large throughput, the system is automated and an analysis software code has been written to provide the user with the masses of several delayed neutron producing isotopes.

2. Theory

When a neutron interacts with ^{235}U the unstable ^{236}U isotope produces fission products and an average of 2.4 neutrons [3]. Immediately following nuclear fission, prompt neutrons are released, accounting for the majority of neutrons produced. Additionally, delayed neutrons are released from some fission products with half-lives ranging from less than a second to almost one minute [4]. The half-lives of these delayed neutrons allow them to be effectively merged into six groups, each with a single nominal half-life and relative yield [3]. Thus, the delayed neutrons can be analyzed to determine the amount of uranium-235 in a sample.

2.1 Mixture Content Determination

If the only fissionable isotope in a sample is uranium-235 the total neutron count provided by the DNCS will be a superposition of the six delayed neutron groups from this isotope. Each delayed neutron group has a different yield per fission; energy and half-life, as shown in Table 1. It is possible to determine the contributions from the individual groups by analysing the delayed neutrons produced as a function of time [5] using equations 1 and 2.

$$Counts_{U-235} = \frac{\varepsilon \nu_{U-235} N_A \sigma_{fission} m \phi}{M_{U-235}} D \qquad (1)$$

Where ε is the efficiency of the detection system, $\nu_{U\text{-}235}$ is the average number of delayed neutrons per fission, determined to be 0.0158 [neutrons fission^{-1}], (the sum of the delayed neutron yields of uranium-235 in Table 1), N_A is Avogadro's number [mol^{-1}], $\sigma_{fission}$ is the thermal fission cross section [m^2] of uranium-235, m is the mass of uranium-235, ϕ is the thermal neutron flux [neutrons cm^{-2} s^{-1}], $M_{U\text{-}235}$ is the atomic mass of uranium-235 [amu] and D is defined as

$$D = \sum_{i=1}^{6} \beta_i \frac{1}{\lambda_i} (1 - e^{-\lambda_i t_{irr}})(e^{-\lambda_i t_{decay}})(1 - e^{-\lambda_i t_{count}}) \qquad (2)$$

Where β_i is the fraction of the total of delayed neutrons for each group, λ_i is decay fraction for each group [s^{-1}], t_{irr} the irradiation time [s], t_{decay} the decay time [s] and t_{count} the count time [s].

Table 1: Delayed Neutron Data for Thermal Fission of ^{235}U [1]

Group	$t_{1/2}$ (s)	λ_i (s^{-1})	Energy (keV)	Yield (neutrons fission^{-1})	βi
1	55.72	0.01	250	0.00052	0.034
2	22.72	0.03	560	0.00346	0.226
3	6.22	0.11	405	0.00310	0.203
4	2.30	0.30	450	0.00624	0.409
5	0.61	1.13	-	0.00182	0.119
6	0.23	3.01	-	0.00066	0.043

Equations 1 and 2 can also be used to determine the masses of other SNM that may be contained in the sample (e.g. ^{233}U and ^{239}Pu) [5]. If a material has *n* delayed neutron producing isotopes, its neutron count after irradiation will be a superposition of the delayed neutrons from each fissile group.

3. Experimental

3.1 Sample Introduction, Irradiation and Analysis

To operate the DNCS the user must place the samples in polyethylene vials which are heat sealed. The vials are then placed in the manual loader. The manual loader is connected to polyethylene tubing through which the samples travel. Once the user loads a sample, compressed air is sent through the tubing, sending the sample into the automatic loader shown schematically in Figure 1. The automatic loader has a perforated metal section, allowing the air to expel, releasing the build up of pressure. The design allows up to 50 samples to be sent to the automatic loader before running the automated program. At this point the user can begin defining set up parameters including the number of samples to be analyzed, irradiation, decay and count time, and the number of times each sample will be irradiated and counted. LabVIEW version 8.6 [6] software then outputs signals to system relays which are in turn wired to all components in the DNCS.

[1] Table adapted from J. Lamarsh *Introduction to Nuclear Engineering 3rd Edition*

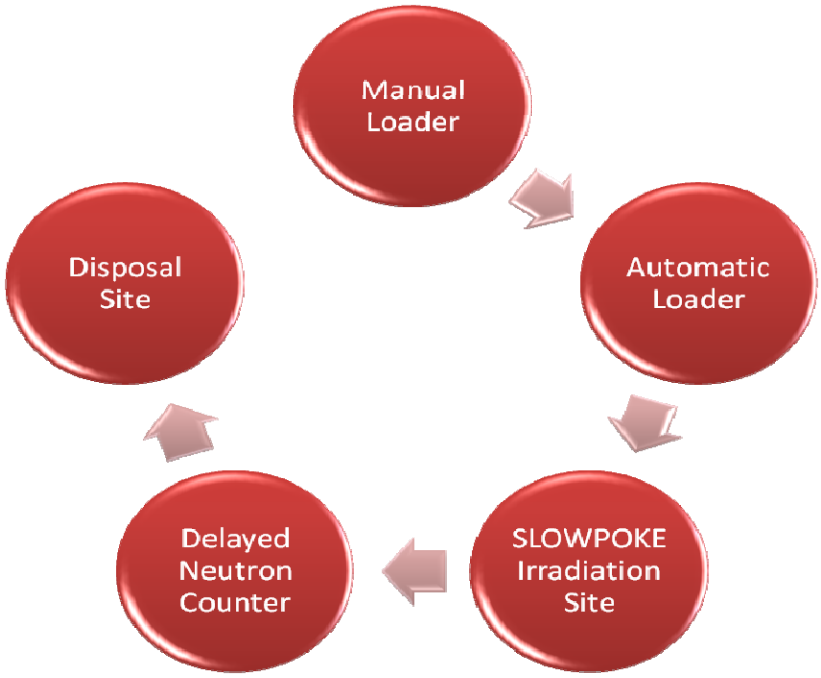

Figure 1: Delayed Neutron Counting System Sample Pathways

Once the LabVIEW program has been started the DNCS loader piston will receive compressed air, this positions the sample over top of the reactor irradiation site. Another valve then pushes air and the sample to an irradiation site, 5.49 m below the reactor pool surface where it is exposed to a typical neutron flux of 5×10^{11} neutrons cm^{-2} s^{-1}. As the sample passes into the SLOWPOKE-2 reactor it will read by a photo-detector indicating to the software that irradiation time should begin. After sitting in the irradiation site for the designated time, the sample is sent up through the loader and diverter into the delayed neutron counter.

The delayed neutron counter (DNC) consists of six equally spaced ^{3}He proportional counters [7] imbedded into paraffin wax surrounding the sample. The wax acts as a neutron moderator, slowing the neutrons down to detectable levels. The six detectors are connected in parallel to a voltage supply and provide information to an Ortec multi-channel analyzer [8] which is read by the LabVIEW software. The sample sits in the middle of the DNC container until the decay time is over, at which point the software begins recording and saving data; including the live time and counts for the user-specified duration. Once the count time has finished, the system has the capability to send the sample either to a disposal site or back into the loader to repeat the process.

The disposal site is located in another area of the DNCS room, sufficiently distant such that negligible contribution to detector background occurs. The sample vial travels through the tubing to this location where it is shielded and has its radiation levels monitored. If the user

would like to collect another run of data, the sample is sent back into the sample feed tubing where the process can begin once again. All of the data are saved onto the computer into a user specified folder. The LabVIEW program produces a text file with counts, energy and time for each sample.

3.2 DNCS Software

3.2.1 LabVIEW Program, Automated Control

The DNCS operation is controlled by LabVIEW data acquisition and control software. Once the LabVIEW executable is opened it provides the user with three operational options, one manual and two automated. The manual option is a useful tool for troubleshooting, it allows the user to turn on each valve manually and check the connections between the relays and USB port. The remaining automated choices for the user are cyclic and pseudo-cyclic. If the cyclic option is chosen, the user can specify how many samples will be analyzed and how many times the irradiation and counting of each sample is to be repeated. If the number of cycles is greater than one, after counting the first time, the sample will be sent to the back of the DNCS sample stack. After the last count of a sample it is sent to the disposal unit. The pseudo-cyclic option is very similar except that after a sample is counted its first time, it skips the sample stack and goes immediately back into the reactor to be irradiated and counted again, after which it goes to the disposal unit.

The LabVIEW DNCS software has two tabs on its front panel, the first is displayed in Figure 2. This display includes a schematic of the DNCS, indicating which valves are on and the position of the sample in the system. The first tab asks the user to define the following properties: manual or cyclic run, number of samples, irradiation, count, decay and cycle delay time, and number of cycles. The second tab of the front panel is a two column live display of time, counts in that interval, and total counts. The final output of the LabVIEW program is an excel spreadsheet containing: sample number, cycle number, time, interval count and count sum.

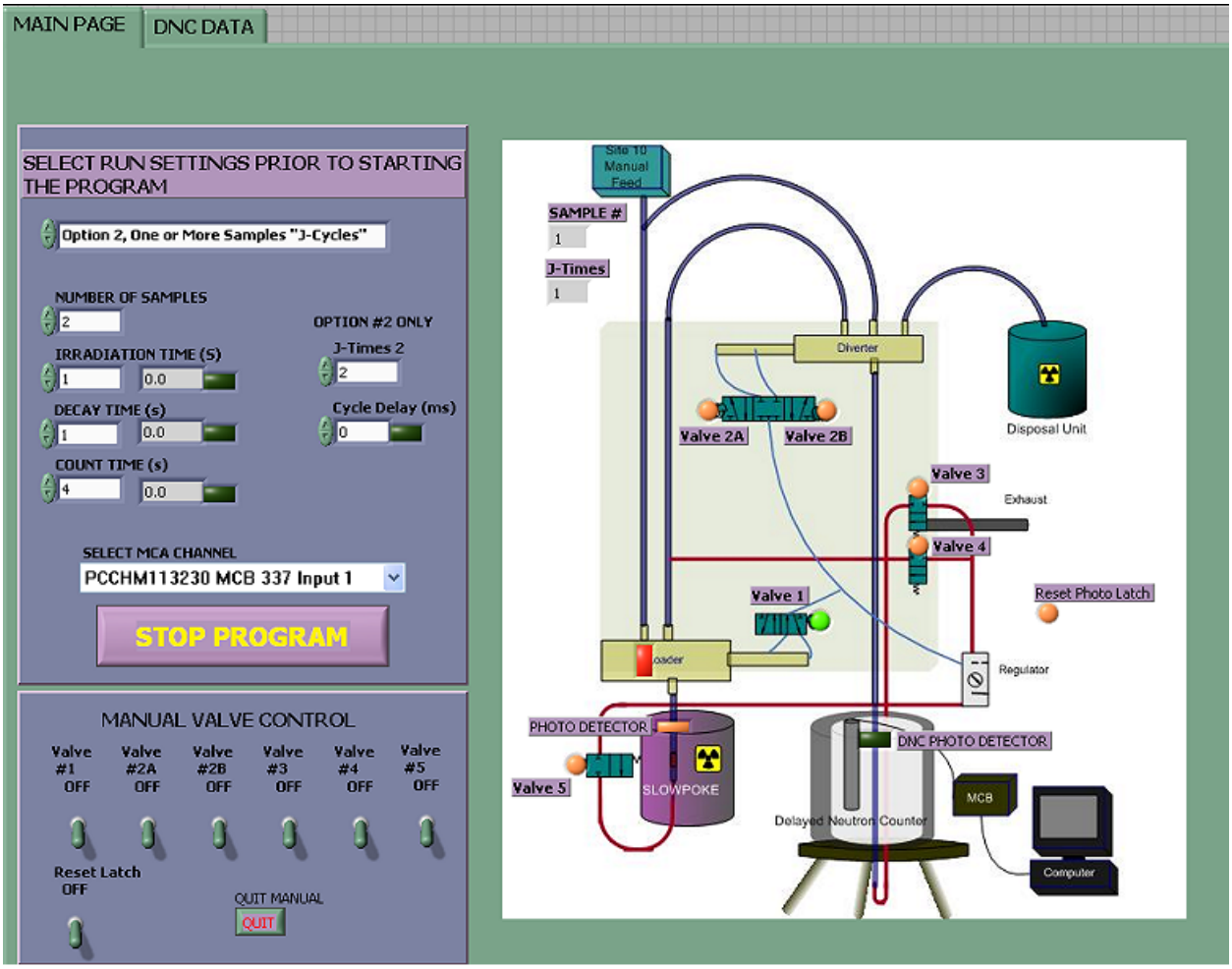

Figure 2: Delayed Neutron Counting System LabVIEW Display

3.2.2 DNCS Deconvolution Program

The DNCS Fissile Analysis Program is a MATLAB R2008b [9] code which takes the output LabVIEW program excel file mentioned in Section 3.1 and determines the fissile isotope masses in each sample. The program examines the time vector and uses equations 1 and 2 to produce a theoretical matrix containing count coefficients for each of the six isotopes for all times involved. The program then manipulates the total count vector and theoretical matrix using Householder's method [10] to determine the fissile isotope masses.

The program prompts the user to input the irradiation time, decay time and number of detectors used. An additional system requirement is that the count time is at least seven seconds, providing the program with enough data to perform the Householder's matrix manipulation. The output format of the fissile analysis program is shown in Table 2.

Table 2: Example Fissile Analysis Results

Sample	Cycle	^{235}U
A	1	$m_A{}^{235}U$
A	2	$m_A{}^{235}U$
B	1	$m_B{}^{235}U$
B	2	$m_B{}^{235}U$

Table 2 shows the output format of the fissile analysis code for two samples counted twice. If there are more data points in the LabVIEW output file than needed to perform the analysis, the code will use these extra values for additional mass determinations. The fissile analysis code examines the LabVIEW output to determine the number of samples and runs. It separates each sample run and determines individual ^{235}U counts. The fissile analysis code is also able to determine the mass of plutonium-239 in the sample, but this has yet to be experimentally verified.

3.3 Experimental Procedure Overview

For the validation procedure the calculated mass of ^{235}U in a sample was compared to the ^{235}U mass measured by the DNCS. For these validation trials, powdered UO_2 samples with a 87.33% purity of natural uranium (obtained from Cameco [11]) where weighed and sealed in separate 7 mL polyethylene vials.

To establish the detector efficiency (ε), a sample powdered UO_2 calibrant containing ^{235}U was first run through the system with an identical neutron flux, number of detectors used, irradiation, decay and count times as the samples with undetermined ^{235}U amounts. These efficiency values were entered into the fissile analysis program which then was utilized to produce the DNCS measured masses of ^{235}U in the trial samples. The same UO_2 was employed for calibrants and undetermined samples, to avoid additional errors associated with uncertainties derived from differences in sample and calibrant purity. The UO_2 was transferred to a pre-weighed polyethylene vial, and re-weighed using a 5 decimal place analytical Mette 601 balance. Trial analytical conditions included modification of the neutron flux (1.00, 3.00, 5.00 x 10^{11} neutrons cm^{-2} s^{-1}), and decay time (5, 25 s).

4. Results and Discussion

As of November 2009, the delayed neutron counting system was installed and successfully irradiated and counted uranium samples. The LabVIEW automation program was completed and tested with all of the DNCS mechanical components. The system is able to process multiple samples at a rate of about one sample per minute. Four amounts UO_2 were prepared and analyzed for their ^{235}U content using the fissile analysis program.

4.1 Results

Four uranium-dioxide amounts were analyzed using the DNCS. Calibrants 1 and 2 were used as standards in order to determine experimental ^{235}U amounts in undetermined samples 3 and 4, respectively. Table 3 shows the calculated ^{235}U amount for each of the samples assuming the UO_2 was composed of 87.33% natural uranium.

Table 3: Calculated Amount of ^{235}U in the UO_2 Samples

Vial Number	Function	^{235}U Mass (μg)
1	Calibration	35.4 ± 0.7
2	Calibration	750.0 ± 0.7
3	Undetermined Sample	68.9 ± 0.7
4	Undetermined Sample	759.0 ± 0.7

All runs had a 5 second irradiation time. The calibration irradiations (vials numbers 1 and 2) were irradiated at three flux levels (1.00, 3.00, 5.00 x 10^{11} neutrons cm^{-2} s^{-1}). Similar flux levels were employed in the analysis of undetermined samples 3 and 4. Calibrant 1 and undetermined sample 3 were analyzed using a 5 second decay time. Calibrant 2 and undetermined sample 4 were counted after a 25 second decay time due to their high ^{235}U contents, which overwhelmed the system electronics after shorter decay times. Table 4 shows a comparison of calculated mass and DNCS measured mass for the undermined materials 3 and 4. This comparison shows that the DNCS measured masses are within experimental error of the calculated values for ^{235}U masses. The four trials with varying fluxes produced an average uranium-235 mass value of 67.5 ± 1.4 and 759 ± 9 µg for samples 3 and 4, respectively (uncertainties obtained using Student's t weighed values at 95% confidence). Both values are in agreement with their calculated values of 68.9 ± 0.7 and 759.0 ± 0.7 µg.

Table 4: DNCS Preliminary Results with Modified Procedure

Sample	Calculated (μg)	DNCS (μg)	Trial Flux ($neutrons\ cm^{-2}\ s^{-1}$)	t_{decay} (s)
3	68.9 ± 0.7	66.7	5.00 x 10^{11}	5
		67.4	1.00 x 10^{11}	
		67.8	3.00 x 10^{11}	
		67.9	5.00 x 10^{11}	
4	759.0 ± 0.7	760.7	5.00 x 10^{11}	25
		754.9	1.00 x 10^{11}	
		762.9	3.00 x 10^{11}	
		759.2	5.00 x 10^{11}	

4.2 Factors Contributing to Experimental Uncertainty

A significant source of error in the ^{235}U determination was found to be time between successive irradiations of a sample. Figure 3 shows the relationship between the amount of time the sample was given to decay after a trial and the error in uranium determination of the subsequent run. Figure 3 contains data for varying samples and ^{235}U amounts. To remedy this problem, the LabVIEW program has a delay option. If samples are counted in the cyclic option more than one time, the user can tell the program to wait a specified amount of time before repeating the process.

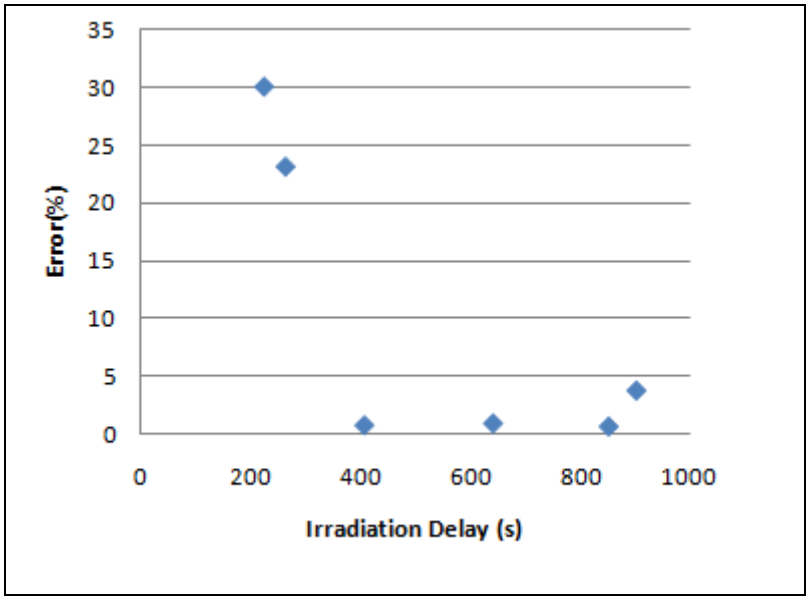

Figure 3: Effects of Delay Time on Experimental Error

An additional contributing factor to experimental uncertainty was the dead time in the DNCS electronics when counting samples with high ^{235}U content. When these samples were sent to the detector a five second delay was not sufficient and the electronics were overwhelmed for over a minute. To eliminate this effect, the samples were irradiated for shorter periods of time, fewer detectors were used, and the user defined decay time was increased.

A sensitivity analysis was performed for all samples to determine the effects of uncertainties in the timings, background contributions, neutron flux and measured mass on the ^{235}U determination. For samples containing smaller amounts of ^{235}U the dominant sources of error include mass measurement error and the irradiation timing. For larger ^{235}U masses, the overwhelming source of error was the irradiation timing. Slight fluctuations in the neutron flux between sample irradiations and the contribution of background counts to the total were found to have little effect on the program's outcome.

5. Conclusions and Future Direction

A DNCS has been designed and installed at RMC to analyze SNM. The DNCS has been successfully validated using powdered samples which contained uranium-235. Further experiments will be conducted with samples taking into account various matrices. In addition, the DNCS will be validated using samples that contain plutonium-239. Finally, experiments will be conducted on SNM samples containing a mixture of fissile isotopes. The results from these experiments will be used to develop a method for the analysis of special nuclear materials at the Royal Military College.

Acknowledgements

The authors would to thank John Shaw for his programming and technical work. In addition, installation and data collection would not have been possible without the help of Dave Ferguson, Kathy Nielsen, Matthew MacKay and Clarence McEwen.

References

[1] M.J. Kristo, D.K. Smith, S. Niemeyer, G.B. Dudder, *Model Action Plan for Nuclear Forensics and Nuclear Attribution,* Lawrence Livermore National Laboratory, 2004.

[2] C.L. Larsson, D.S. Haslip, *DRDC Consolidated Canada Results to the HEU Round Robin Exercise,* Defence Research and Development Canada, 2004.

[3] J. Lamarsh, *Introduction to Nuclear Engineering Third Edition,* Prentice-Hall, 2001.

[4] G.R. Keepin, T.F. Wimett, R.K. Zeigler, *Delayed neutrons from fissionable isotopes of uranium, plutonium, and thorium,* Journal of Physical Review 107 (1957) 1044.

[5] X. Li, R. Henkelmann, F. Baumgartner, *Rapid Determination of uranium and plutonium content in mixtures through measurement of the intensity-time curve of delayed neutrons,* Journal of Nuclear Instruments and Methods in Physics Research B 215 (2004) 246.

[6] National Instruments, *User Guide and Specifications NI USB-6525*. National Instruments Corporation.

[7] Reuter Stokes, *Reuter Stokes He-3 Proportional Counter* RS-P4-0406-212*,* GE Energy.

[8] EG&G ORTEC *Model 919 Spectrum Master™ Hardware Manual*, ORTEC.

[9] The Mathworks, *Matlab 7 Programming Fundamentals,* COPYRIGHT 1984–2009 by The MathWorks™, Inc.

[10] A. Householder, *Principles of Numerical Analysis,* Dover Publications 1953.

[11] Cameco, Uranium Dioxide Powered Sample, Lot Number 2005232, 2005.