An Examination of the Time-Dependent Background Counts of the Delayed Neutron Counting System at the Royal Military College of Canada

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Abstract

A delayed neutron counting (DNC) system for the analysis of special nuclear materials (SNM) has been constructed and calibrated at the Royal Military College of Canada. The polyethylene vials used to transport SNM samples have been found to contribute a time-dependent count rate, *B(t)*, far above the system background. *B(t)* has been found to be independent of polyethylene mass and shows a dependence on irradiation position in the SLOWPOKE-2 reactor and irradiation time. A comparison of *B(t)* and the theoretical delayed neutron production from the fission of small amounts of ^{235}U has indicated that trace amounts of uranium may be present in the DNC system tubing.

1. Introduction

The Royal Military College of Canada (RMC) has developed and installed a delayed neutron counting (DNC) system within the SLOWPOKE-2 Facility at RMC with the objective of enhancing the special nuclear materials (SNM) analysis capabilities of the Facility [1]. DNC is an efficient and non-destructive technique capable of determining the mass of SNM nuclides including ^{235}U, ^{239}Pu and ^{233}U over a large range of concentrations. In this technique unknown samples containing fissile nuclides are irradiated by a neutron source and the subsequent delayed neutrons that are produced are counted by neutron detectors. The counts from the unknown sample are compared to calibration standards to determine the fissile content. Traditional DNC systems record the cumulative delayed neutron count from unknown samples, which is sufficient when analyzing samples with one known fissile isotope. The system at RMC is based on recent work [2] which has shown that the fissile content of samples containing two fissile isotopes can be determined by the analysis of the time-dependent neutron count rate.

The DNC system at RMC records and analyzes the time dependent count rate of the delayed neutrons produced by the irradiation of ^{235}U, ^{239}Pu and ^{233}U and has been validated for ^{235}U using certified reference materials in a manner that is consistent with ISO 17025 standards [1,3]. It was noted early in the validation process that the polyethylene (PE) encapsulation vials contribute to the overall neutron count rate recorded by the system detectors. These apparent neutron emissions from the polyethylene vials have been examined by others including Benzing *et al*. [4], who recorded the cumulative counts from the polyethylene vials and were unable to establish their origin. The present work describes the investigation of the time

dependent count rate resulting from background, the irradiation of vials, and the inferred contribution of the DNC transport system. The capability at RMC to record neutron rate in 1s intervals provided the opportunity to examine the time dependent neutron background in great detail.

2. Theory

The SNM nuclides ^{235}U, ^{239}Pu and ^{233}U will release prompt neutrons and fission fragments upon interaction with thermal neutrons. Delayed neutrons are produced when some of the fission fragments undergo β^- decay. The half-lives of delayed neutrons range from less than one second to almost one minute [5]. These neutrons are sorted by half life into eight groups with associated production ratios [6] shown in Table 1 where ν_i is the production of delayed neutrons from group i, ν_d the total number of delayed neutrons and ν_t all neutrons produced in the thermal fission of ^{235}U.

Table 1: Delayed Neutron Data for the Thermal Fission of ^{235}U.

Group	$t_{1/2}$ [s]	λ [s^{-1}]	$\beta_i = \nu_i/\nu_d$ [%]	$\beta_i=\nu_i/\nu_t$ [%]
1	55.6	0.014267	0.0328 ± 0.0042	0.0218 ± 0.0029
2	24.5	0.028292	0.1539 ± 0.0068	0.1023 ± 0.0036
3	16.3	0.042524	0.091 ± 0.009	0.0605 ± 0.0063
4	5.21	0.133042	0.197 ± 0.023	0.131± 0.016
5	2.37	0.292467	0.3308 ± 0.0066	0.2200 ± 0.0083
6	1.04	0.666488	0.0906 ± 0.0046	0.0600 ± 0.0036
7	0.424	1.634781	0.0812 ± 0.0016	0.0540 ± 0.0021
8	0.198	3.554600	0.0229 ± 0.0095	0.0152 ± 0.0064
Total	**9.020**	**0.076849**	**1.000 ± 0.029**	**0.665 ± 0.021**

The rate of delayed neutron counts for an nuclide j, $S(t)_j$, is described by

$$S(t)_j = m_j\left(\frac{\varepsilon \nu_j N_A \phi \sigma_{f_j}}{M_j}\right)\sum_{i=1}^{k}\beta_{ij}\left(1-e^{-\lambda_i t_{irr}}\right)\left(e^{-\lambda_i t_d}\right)\left(e^{-\lambda_i t}\right) \quad (1)$$

where ε is the detector arrangement efficiency, ν_j is the average number of delayed neutrons emitted per fission, σ_{fj} is the fission cross section (b), N_A is Avogadro's number (mol^{-1}), ϕ the neutron flux (cm^{-2} s^{-1}), M_j is the atomic mass number (g mol^{-1}), k is the total number of delayed neutron groups, β_{ij} is the fraction of total delayed neutrons for group i, λ_i is the decay constant for group i (s^{-1}), t_{irr} is the irradiation time (s), t_d is the decay time (s), t is the time post decay (s) and m_j is the fissile mass (g) [2].

It has been established that the DNC system at RMC displays both a time independent and a more substantial time dependent background contribution [1]. The count rate, *C(t)*, of the DNC at RMC can therefore be expressed as;

$$C(t)_j = S(t)_j + B(t) + Background \quad (2)$$

where *B(t)* is the time dependent background correction for the vials.

3. Experimental

Typical use of the DNC system requires samples to be heat sealed in a 1.5 mL polyethylene vial (LA Packaging, Yorba Linda, CA) which is then sealed in a 7.0 mL vial with another 1.5 mL vial occupying the remaining void. Vials contain Al, V, Na, S, Mg, Br, Cl, Mn, I, K and Co impurities at concentration of <2 ppb [7]. None of the identified impurities are known to produce delayed neutrons upon irradiation in a thermal neutron flux. Uranium samples were prepared from a CRM stock solution containing 7.32 ppm ^{235}U.

The DNC system at RMC is controlled with a custom created LabVIEW (2009, National Instruments, Austin Texas) based graphical user interface. The experimental parameters that can be customized include irradiation time, decay time before counting, count time and the number of samples to be analyzed. Once the parameters have been defined, the DNC system sends the samples through the irradiation-decay-count procedure.

Typical DNC system operation consists of sending the samples to an inner site of the SLOWPOKE-2 reactor where the thermal neutron flux is 5.5 x 10^{11} $cm^{-2}s^{-1}$ (±5%) at half power [8]. Once a sample has been irradiated for the designated time it is sent to the centre of the neutron counter which contains six ^{3}He proportional detectors (RS-P4-1613-202, GE Energy, Twinsburg, OH) embedded in paraffin wax in a hexagonal arrangement. Neutron counts are provided by a preamplifer (ORTEC 142, Oak Ridge, TN), voltage supply (ORTEC 556), and amplifier (ORTEC 575A) and a multichannel buffer (ORTEC 919E) before the samples are sent to an isolated disposal bin located approximately 5 m from the apparatus.

The information collected by the LabVIEW is exported as MS Excel™ file and analyzed using a MatLab™ (R2008a, Mathworks, Natick, MA) code developed at RMC, which is used to calculate the amount of ^{235}U in each sample. Reference [1] contains a detailed description of DNC system hardware and software.

4. Results & Discussion

The experiments discussed in this section were designed to examine the source of background counts in the DNC system. The three sources considered include: (i) the background counts, (ii) the polyethylene vial material, and (iii) contaminants within the DNC sample transport system.

4.1 DNC system background counts

The system was set for an automatic 60s count duration to measure the background of the system in the absence of the polyethylene vials with the reactor operating at half power. No samples were placed in the loader and the system proceeded to record the neutron count rates of the reactor air, which contains ^{41}Ar and ^{4}He among other nuclides, pushed through the pneumatic tubing into the counter arrangement. The count rate was recorded in 1s intervals for the duration of the count time. This procedure was repeated eight times and the background counts of the system showed no dependence on count time. This implied the radionuclides which may have been present in the gas stream coming from the SLOWPOKE-2 were not contributing to the overall delayed neutron count. The system background in the absence of a vial was determined to be 3.8 ± 0.4 counts per second. The count rate distribution for the 1s recording intervals is illustrated in Figure 1 for the eight trials.

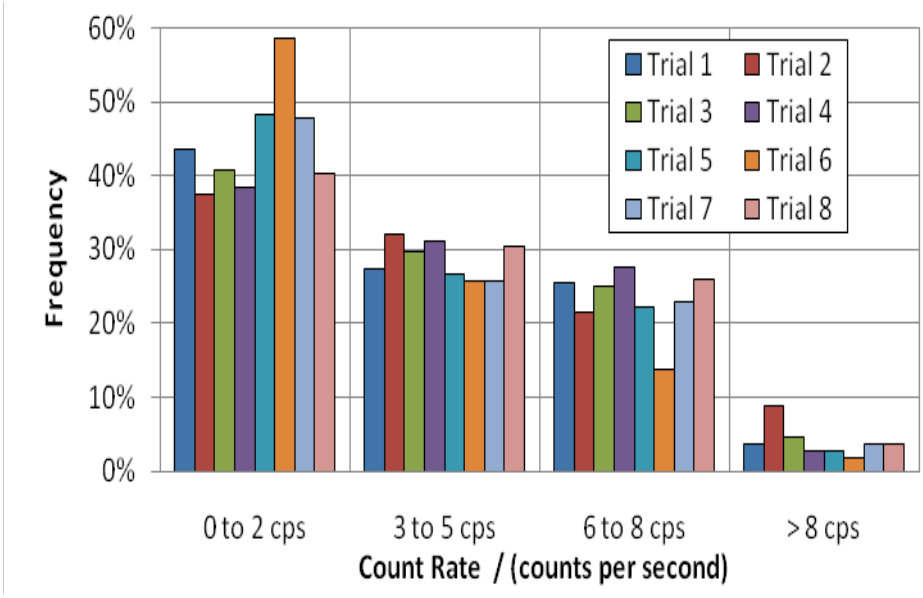

Figure 1: Background count rates for the DNCS system

4.2 Polyethylene vial background

4.2.1 Consistency of *B(t)*

Eight polyethylene vials were prepared in a manner similar to typical use to examine the any SNM sample (two inner 1.5 mL PE vials sealed inside a 7.0 mL vial). Each sample was then irradiated for 60s with the SLOWPOKE-2 operating at half power. The neutron count rate was recorded after the sample was irradiated. Figure 2 shows the count rates for eight trials. *B(t)* remained statistically consistent for each individual vial and over a period of several days. A comparison of the background count rate (3.8 ± 0.4 cps, Section 4.1) and the vial count rates, in Figure 2, confirm a time dependent count rate resultant from the presence of the vials.

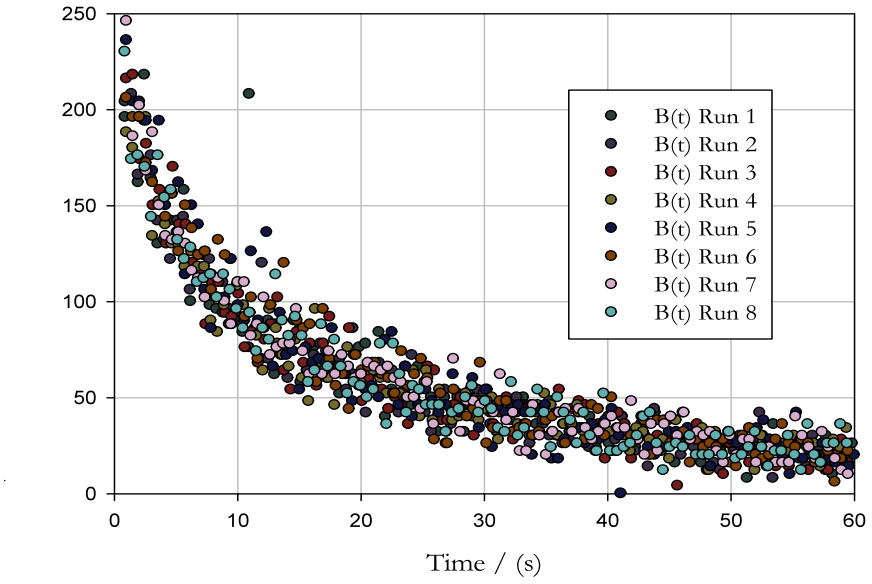

Figure 2: The time-dependent count rate for eight polyethylene vial trials.

4.2.2 Count rate independence of polyethylene mass

To examine the possibility of the activation of impurities in the polyethylene, twenty four vials were prepared with varying total mass of polyethylene. Eight vials were left empty, eight contained two inner 1.5 mL vials, and the remaining eight were filled with pieces of additional polyethylene (produced by cutting the vials into small pieces). The total polyethylene mass ranged from 3.3 to 6.6 g. The total mass of polyethylene had no apparent effect on *B(t)*, Figure 3, indicating that the source of the delayed neutrons maybe from a contaminant deposited from the DNC transport system onto the vials.

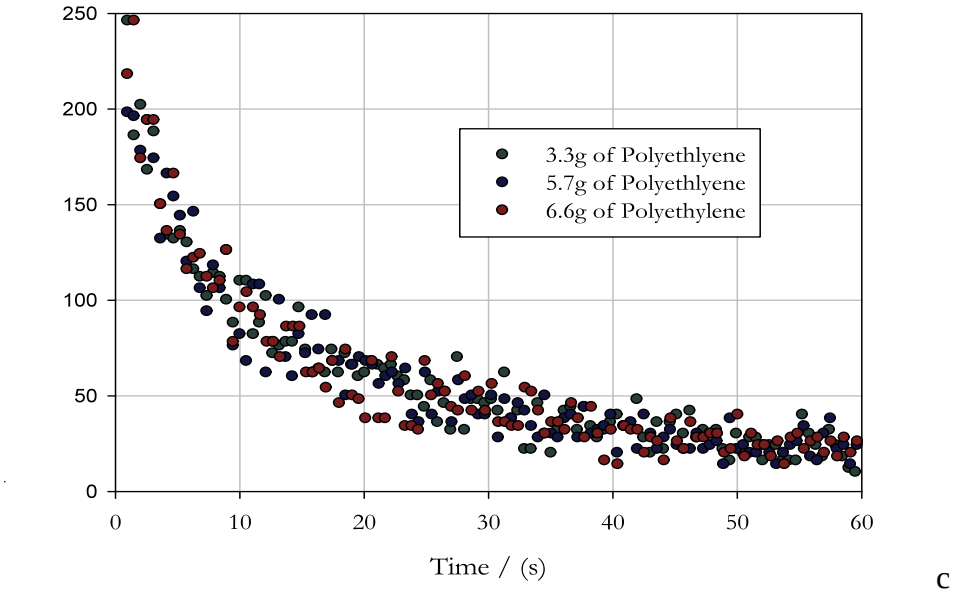

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Figure 3: *B(t)* for polyethylene vials of varying total mass

4.3 Contaminants within the DNC sample transport system

4.3.1 Irradiation site comparisons

The polyethylene mass independence of *B(t)* indicates the vials may be picking up a substance containing delayed neutron generating nuclides during its travel through the DNC system. The original DNC system site (5) was temporarily switched to another inner irradiation site in the SLOWPOKE-2 (site 2) to examine any possible effects the site location may have on *B(t)*. The background count rate was determined in an identical manner as Section 4.1 and is compared in Figure 4. The background count rates in sites 2 and 5 were found to be indistinguishable.

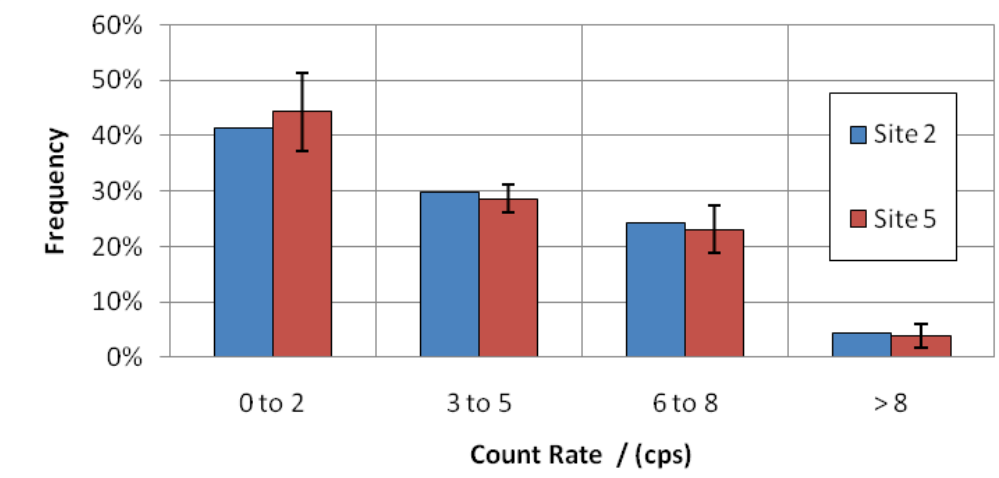

Figure 4: Background count rates for SLOWPOKE-2 sites 2 and 5 (with 1σ uncertainty)

Twenty new vials were prepared in the manner described in Section 4.2.2 and irradiated for 60s in site 2. After the vials had been irradiated in site 2 the resulting count rate was found to be consistent with the DNC system's background counts, 3.8 ± 0.4 and 3.9 counts per second, respectively. Figure 5 illustrates the comparison of *B(t)* for SLOWPOKE-2 sites 2 and 5 for the background, 3.3 g PE vial count rates and 6.6 g PE count rates. All vials irradiated in site 2 were found not to contribute additional counts to the DNC system background. The increased count rates from the polyethylene vials irradiated in site 5 indicates the presences of a delayed neutron generating nuclides contaminant in that irradiation site.

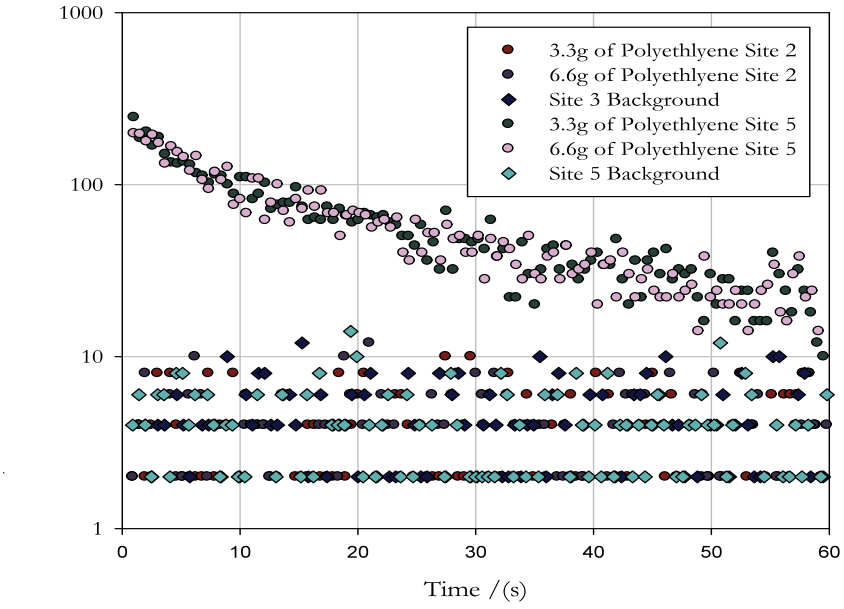

Figure 5: A comparison of *B(t)* for SLOWPOKE-2 sites 2 and 5

4.3.2 Contaminant identification

To identify the quantity and type of nuclide contaminants, several samples containing small amounts of ^{235}U were analyzed by the DNC system. The vial and background contribution to count rates in Eq. (2) were subtracted and the resulting *S(t)* was compared to *B(t)*, Figure 6. As illustrated in Figure 6, the similar trending in the *B(t)* curve, indicates that the source of contamination is ~100 ng of ^{235}U.

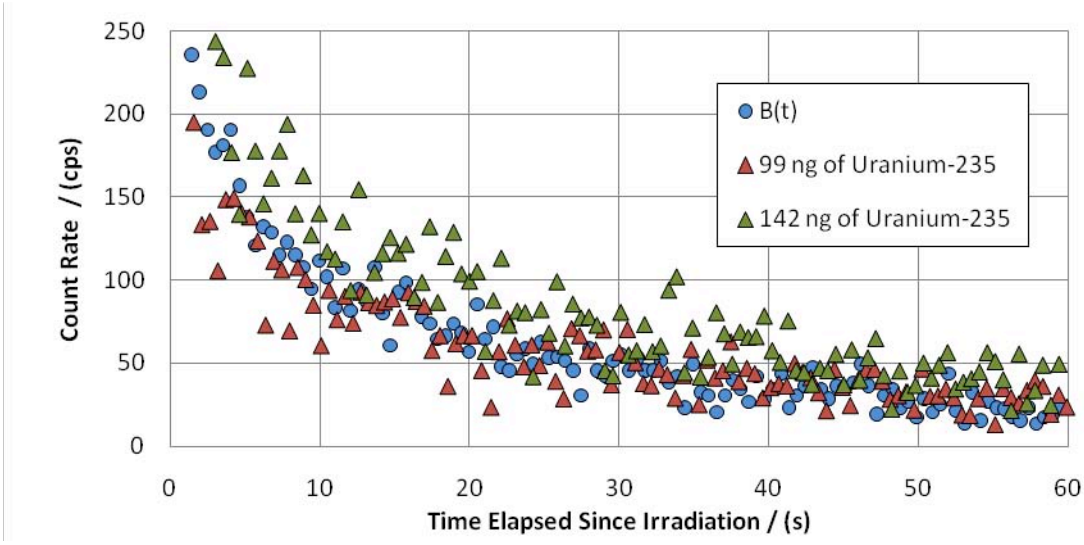

Figure 6: Comparison of *B(t)* to *S(t)* for small amounts of ^{235}U

Further experimentation for varied irradiation times, 10s and 60s, was completed using two 7.0 mL polyethylene vials (each containing two smaller 1.5 mL vials). The results, Figure 7, indicate that recorded delayed neutron count rate varied with irradiation time. This suggests that the ^{235}U contamination is from the site 5 transport system and not from within site 5 as ^{235}U within the site would have been irradiated continuously and the recorded count rate from such a sample would not vary with irradiation time.

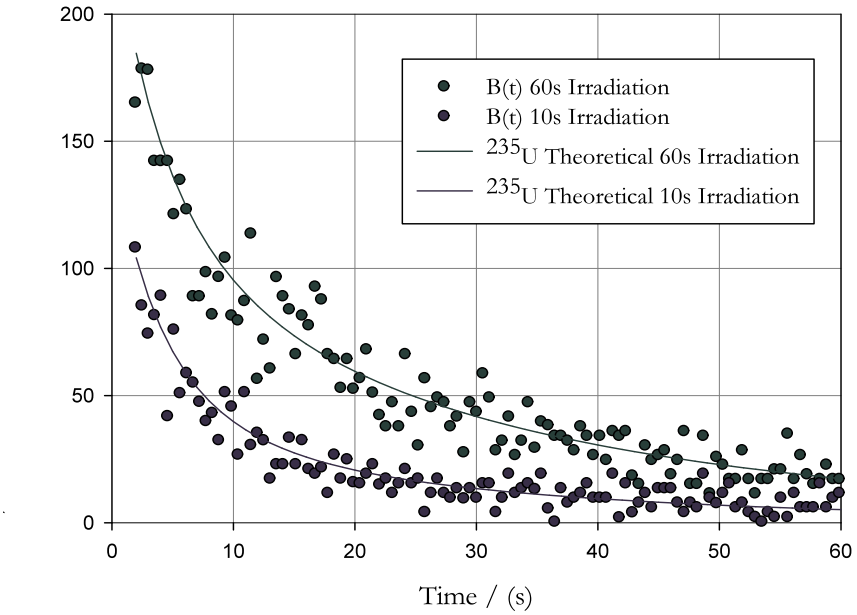

Figure 7: 10s and 60s irradiations, theoretical and *B(t)*.

4.4 Additional considerations

Other institutions with DNC systems have incorporated lead shielding into their systems to reduce gamma-ray background [2]. A multi-element gamma ray source was used to examine the possibility of *B(t)* being produced by gamma-ray interference. The source (with gamma energies in the range of 46 – 1836 keV) was first placed on top of the counter arrangement and finally held in direct contact with a ^{3}He detector each of which resulted in a count rate indistinguishable from background. The contribution to the delayed neutrons count rate from fast neutron reaction ^{17}O(n,p)^{17}N with half life of 4.17 s is also assumed to be small [9].

5. Conclusion

The contribution of the polyethylene vials, *B(t)*, was found to be characteristic of the contaminant within the DNC transport system of site 5. The source of contamination has been identified as ~100 ng of ^{235}U. Further work to remove this contamination from the DNC transport system will be an ongoing effort of this research. Inductively coupled plasma mass spectrometry will also be used to identify the nuclides present on the vial surface after sample analysis through delayed neutron counting.

6. Acknowledgements

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7. References

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