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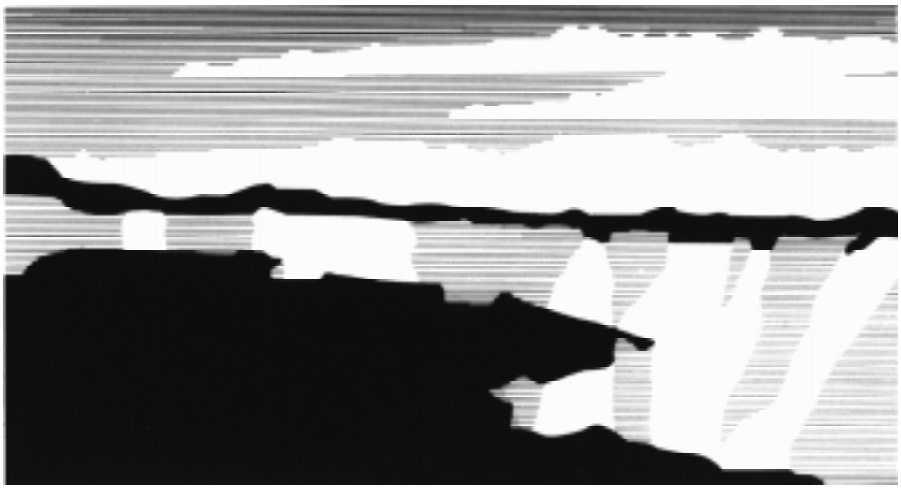

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Measurements of Absolute Delayed Neutron Yield and Group Constants in the Fast Fission of U-235 and Np-237

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The delayed neutron activity resulting from the fast induced fission of U-235 and Np-237 has been studied. The decay constants, relative abundances, and absolute yield of delayed neutrons from fast fission of U-235 and Np-237 were measured using the Godiva IV fast assembly at the Los Alamos Critical Experiments Facility. The absolute yield measured for U-235 was 0.0163 ± 0.0008 neutrons/fission. This value compares very well with the well-established Keepin absolute yield of 0.0165± 0.0005. The absolute yield value measured for Np-237 was 0.0126 ± 0.0007. The measured delayed neutron parameters for U-235 are corroborated with period (e-folding time) vs reactivity calculations.

1. Introduction

Since their discovery, many of the properties of delayed neutrons have been extensively studied. These quantities, essential to the understanding of reactor kinetics, have also gained more importance in other fields such as fissile material assay or neutron flux monitoring for fissionable materials. Accurate understanding and measurement of delayed neutron yield data are essential for fast reactor systems. Until quite recently the U-235 delayed neutron parameters measured by Keepin et al.[1] have been recommended as the "best" values; however, some new theoretically derived delayed neutron parameters[2] have showed considerable differences when used in reactivity calculations.[3,4,5]

There are several reasons for the interest in Np-237. Those who evaluate the very long term risks associated with the disposal of radioactive wastes have been very concerned with Np-237. This concern is based on (a) its presence in high-level waste from the nuclear fuel cycle, (b) its very long-half-life (2×10^6 years), and (c) its critical mass. Spent nuclear fuel is dominated by higher-mass actinides such as Np-237, and the safe geological disposal of these isotopes, which have long-lived decay chains, is difficult to prove. Neptunium-237 dominates the toxicity of highly active waste after 10,000 years.

Alternatives have been proposed to dispose of these actinides. Under consideration is a neptunium burner.[6] Neptunium could be separated during reprocessing and subsequently used as fuel in a sodium-cooled fast reactor that has a very hard neutron spectrum. Studies show that one reactor of this type could suffice to convert into fission products the neptunium produced by 125 light water reactors. Therefore, for the safe operation of such a reactor, which would have large quantities of higher actinide isotopes in its core, a thorough knowledge and understanding of each actinide, such as delayed neutron data, is essential.

In general the knowledge of delayed neutrons for U-235 prior to this experiment was satisfactory for most practical applications. However, because of recent reactivity calculations[3,4,5] showing differences from the time-honored Keepin values justified a further examination of the delayed neutron data. The six-group representation of Np-237 lacks experimental determination. The improved technology, which includes better detector type and geometry, and a new data acquisition system, provides a better means to examine existing and new delayed neutron parameters.

2. Detection System and Data Acquisition

The neutron detector must meet a number of requirements. First, it has been shown that the count rate decreases by three orders of magnitude in about 100 seconds[1]; therefore, a detector with adequate neutron efficiency but with low dead-time is needed. The detector must also be insensitive to gamma rays. The detector efficiency should also be insensitive to small sample displacement. A detector that meets these requirements is the well counter described by Anderson et al.[7] The well counter consists of 20 He-3 tubes embedded in a cylindrical configuration inside polyethylene. The counter is surrounded by 10 cm of polyethylene, lined with a 1.5-mm-thick sheet of cadmium. The layer of cadmium is intended to absorb background thermal neutrons that are created outside the cavity. To further reduce the high neutron background from Godiva IV, two additional layers of neutron absorbers have been placed around the well counter. The first layer is a 2-cm-thick sheet of Boral. Boral is a uniform dispersion of boron carbide with aluminum. The second layer consists of 7.6-cm-thick borated polyethylene. This shielding was sufficient to reduce background neutrons from Godiva to negligible levels. Figure 1 shows the cross sectional view of the well counter.

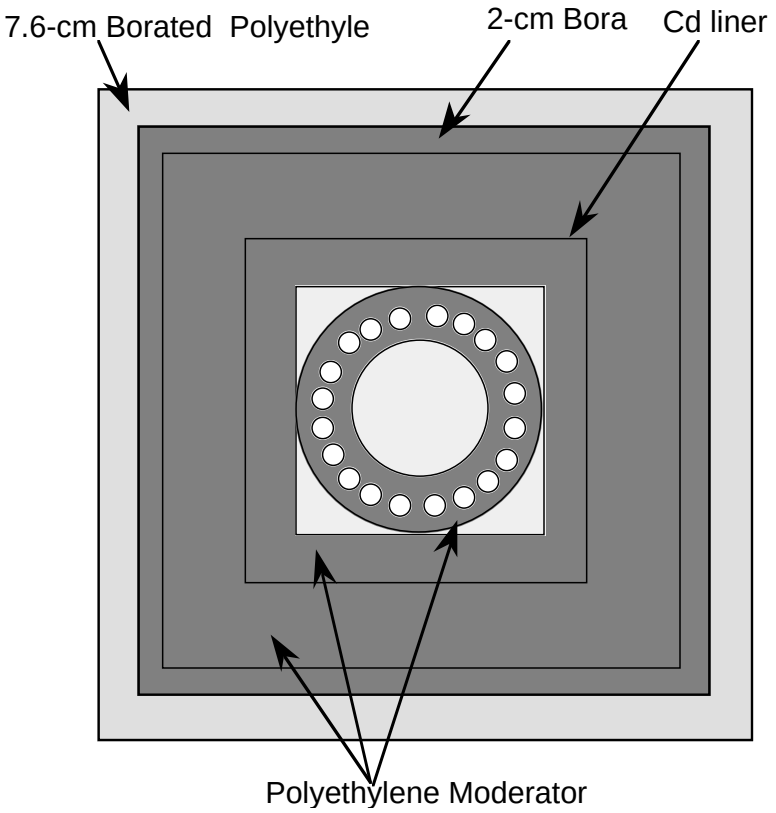

Figure 1. Cross Sectional View of the Well Counter.

The He-3 tubes have an active length of 25 cm and outside diameter of 2.54 cm. The tubes are 98% filled with He-3 at a pressure of 4 atm. The twenty He-3 detector tubes are arranged as 5 banks of 4 counters each, making the detector a segmented device consisting of 5 segments. This arrangement was used to recognize spurious data that might occur as a result of misbehavior of one tube bank, and to improve the effective dead time.

The electronic system for the measurement of delayed neutrons consists of the following equipment. The four detectors in each counter bank were summed by Tennelec TC-175 preamps, and the output from the preamps was processed by Tennelec 202BLR amplifiers and LeCroy 623B discriminators. The processing module, a LeCroy 429A Fan in/Fan out, records the total neutron counts per counting interval and stores the total number of neutron counts detected by each of the five detector banks in an Ortec 772 Scaler. The output signals from the neutron detector discriminators are also fed into the input synchronizer of the Pulse Arrival-Time Recording Module (PATRM). The PATRM, with accompanying software, records the arrival times of pulses from the detectors in a sequential manner. Current techniques convert the raw time

history data into other data domains such as (a) the number of pulses in a gated window or (b) the number of pulses in a shift register after a pulse exits. So, the original pulse arrival-time history is completely lost and the data analysis is limited to one or two algorithms. These current methods have their parameters (such as gate width, clocks, shift-register lengths, etc.) set before the data collection in the hardware or software. Therefore, once the data is taken, any change in the parameters requires running the experiment again. By using the PATRM, the original pulse arrival-time history can be recorded and saved. Thus, one can apply many algorithms to the same data and also change the parameters (gate width, bin width) used in the analyses. This method of recording the original data also allows the user to use newly developed algorithms to analyze data taken previously.[8]

The absolute efficiency of the well counter was determined by using a newly calibrated Am/Li source. This type of source was selected because its energy spectrum is very similar to that of delayed neutrons.[9] The absolute efficiency calculated in the well counter is 29.04% ± 0.6%. The dead-time in the counting system was measured by two techniques. The first technique used the PATRM module to observe the dead time,[10] while the second technique used the two source method. The dead-time calculated by these methods was 0.46 ± 0.02 μsec. As previously mentioned the detector efficiency should also be insensitive to small sample displacement. To investigate this effect, the efficiency of the detector was measured as a function of axial distance from the bottom of the detector cavity. An Am/Li source was placed at the bottom of the cavity and moved axially upwards with the detector system in the vertical position. The efficiency of the detector as a function of neutron-source position was constant through a displacement range of 10 cm. Therefore, the irradiated sample can be stopped at any point within this 10 cm range, and the response will be constant.

3. Transfer Apparatus

To analyze the delayed neutron decay, it is necessary to transfer the samples from a region of high flux to a well-shielded counting area. The transfer system or “rabbit” fulfills this condition. The rabbit system is an electronically controlled pneumatic transfer device. The rabbit system moves the sample from the irradiation position near Godiva IV to the counting position inside the detector in 110 msec. Once the delayed neutrons have been measured in the well counter, the sample is retrieved and a cycle is completed. The sample is retrieved to its irradiation position by the use of a fishing reel. The retrieval system works by attaching a fishing line to the end of the capsule and uses a fishing reel to retrieve the sample. The total distance traveled by the sample is 4.52 meters. The general experimental setup with the transfer system is shown in Figure 2.

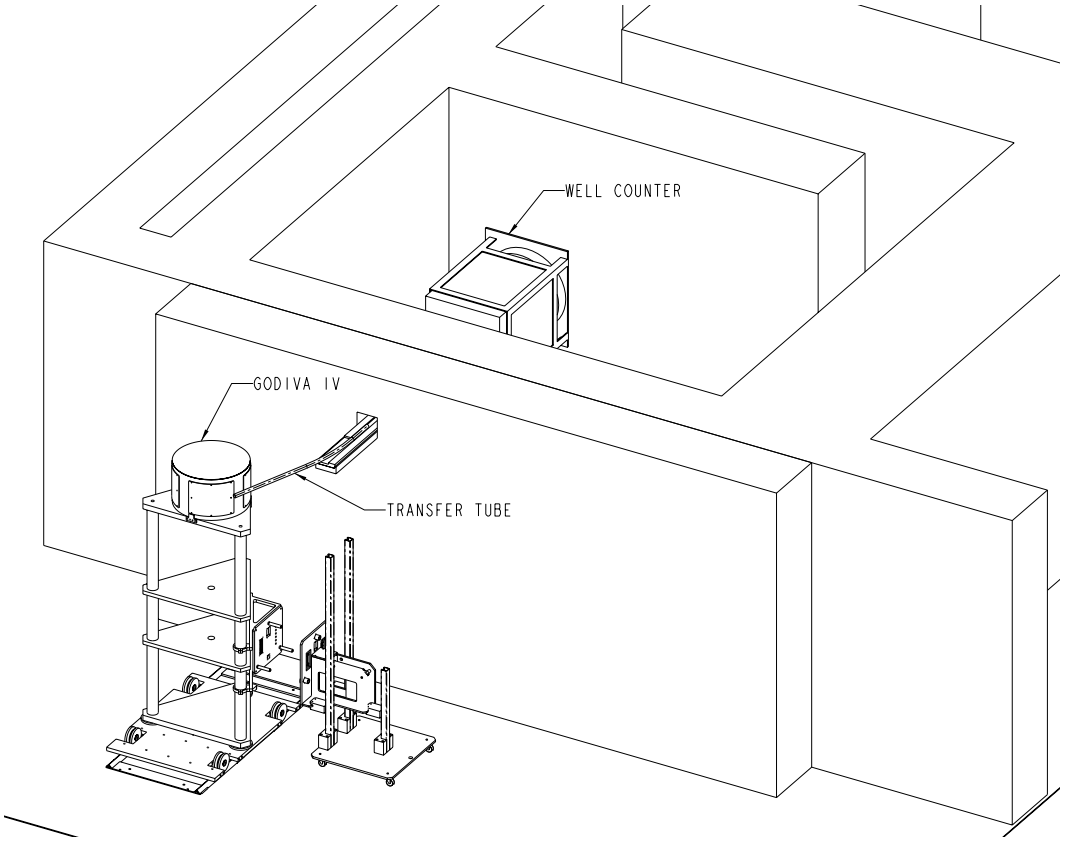

Figure 2. Schematic diagram of experimental setup with Godiva and the counter.

The data-collection scheme entails irradiating a small sample of U-235 or Np-237 with the Godiva IV burst critical assembly. The sample is irradiated over a period of time to accentuate particular delayed neutron

groups. Irradiations can be long to saturate the population of delayed neutron precursors, or short (burst) to accentuate the short-lived groups. After irradiation, the sample is transferred to its counting position and this transfer is initiated by a trigger signal obtained from a photomultiplier placed close to Godiva. The delayed neutrons are counted until count rate diminishes to background levels (1-2 cts/sec). The software collects the data and starts the clock in the PATRM when the output of the photomultiplier is activated by the burst. In this manner the departure or initial time is linked to a reproducible event. However, the PATRM is not enabled to collect data until an infrared diode inside the detector senses the arrival of the sample. This sequence avoids acquisition of counts that are not associated with the sample. The software scans the list with bin widths of 20 msec at first, then the time bin is doubled every 50 bins. The reason for varying the size of the time bins is to better observe the complicated decay of delayed neutrons. At the beginning of the decay, the delayed neutron precursors include those with short half-lives. To better fit this data, small time bins are applied. As the decay continues, the half-lives of the precursors are longer, and the bin width is adjusted accordingly.

The U-235 and Np-237 samples are placed inside a polycarbon capsule to facilitate the transfer of samples and minimize damage to samples. The sample masses were selected to compromise between low counting rates for small masses and excessive multiplication effects for the larger masses.[11] To ensure consisting efficiency, it is required that the sample be stopped at the same position. Several types of dash pots were tried to stop the slug. Ultimately, a piece of nylon rope was used to absorb the impact. This rope absorbed and reduced the bounce to less than 0.5 cm.

4. Analysis of Delayed Neutron Parameters

The complicated delayed neutron activity can be best measured when the irradiation time of the sample is (a) instantaneous, short compared to the shortest delayed neutron period or (b) infinite, long compared to the longest delayed neutron period. The infinite irradiation refers to delayed critical operation of Godiva IV, and the instantaneous irradiation is obtained by small pulses from Godiva that have a FHWM of 22.6 msec duration. Both types of irradiation were adjusted to produce tolerable count rates in the detector system. The four longer periods and their abundances are calculated from the infinite irradiation while the four shorter periods and their abundances are calculated from the instantaneous irradiation. The parameters for groups 3 and 4 are calculated by combining data from the instantaneous and saturated irradiations.

The analyses of the delayed neutron activity are modeled with the assumption that the activity as a function of time can be represented by a linear superposition of an exponential decay period. Thus the decay of the delayed neutron activity for the saturation irradiation can be represented by

$$y_S(t) = \sum_{i=1}^{M} a_i e^{-\lambda_i t} \tag{1}$$

while the model for the instantaneous irradiation is represented by

$$y_I(t) = \sum_{i=1}^{M} a_i \lambda_i e^{-\lambda_i t} \tag{2}$$

where y is the counting rate, t is the independent variable time, a_i is the abundance, and λ_i is the decay constant of the *i*th group. Equation (2) uses an effective weighting of the initial activity by the factor of λ_i. This is advantageous in studying the short-lived delayed neutron groups. The summation is taken over the necessary number of groups for good convergence of the delayed neutron groups. This experiment takes the summation over six groups, keeping the tradition established by Keepin et al.[1] To determine how well the model agrees with the data, we use the chi-square function.[12] If we assume in equations (1) and (2) that a_{io} and λ_{io} are the initial estimates of the "real" parameters a_i and λ_i, we can then proceed to use an iterative nonlinear least squares method to find the final values a_i and λ_i which will minimize the chi-square function. The iterative Levenberg-Marquardt[13] method is used to solve for the unknown delayed neutron parameters simultaneously.

Most algorithms for the least-squares estimation of nonlinear parameters have centered about either of two approaches, the Gauss-Newton method and the steepest-descent method. Marquardt[13] introduced an algorithm that combines the two methods. His method is based on early work of Levenberg.[14] The Levenberg-Marquardt technique performs an optimum interpolation between the two methods by introducing a constant that is varied to accelerate the convergence. Equation (3) shows a representative matrix equations to illustrate the Levenberg-Marquardt technique where a constant φ has been introduced.

$$\left(A^{(r)} + \varphi^{(r)}\, I\right)\delta a = b^{(r)} \tag{3}$$

δa is the step that is being taken to solve at the *r*th iteration, I is the unity matrix and φ is a positive constant that is controlled during the iteration. When the constant $\varphi \rightarrow 0$, equation (3) approaches the steepest-descent method and when $\varphi \rightarrow \infty$, equation (3) approaches the Gauss-Newton method. This hybrid method works very well, and is used to solve for the unknown parameters in equations (1 and 2). The method is implemented in a Mathcad subroutine which uses the public domain MINPACK algorithms developed and published by the Argonne National Laboratory.[15]

5. Absolute Delayed Neutron Yield and Fission Rate Measurements.

The most accurate technique to calculate the absolute yield is to observe the total number of counts following an instantaneous irradiation where the initial activity is proportional to $a_i \lambda_i$ as shown in equation 4.

$$C_{tot} = \varepsilon F_s Y_{abs} \int_0^{\infty} \sum_i a_i \lambda_i e^{-\lambda_i t} dt = \varepsilon F_s Y_{abs} \tag{4}$$

Where C_{tot} is the total count rate, ε is the absolute efficiency of the detector, F_s is the total number of fissions, and Y_{abs} is the absolute delayed neutron yield. Therefore, the absolute yield is calculated directly from the total counts as:

$$Y_{abs} = \frac{C_{tot}}{\varepsilon F_s} \tag{5}$$

The total number of fissions, F_s, produced in a sample during a Godiva irradiation was measured by a standard foil-activation technique. For this experiment the activities of Ba-140, La-140, and Mo-99 were observed. These fission products have relatively long half-lives which permit the measurement of their activities in a clean gamma spectrum after the short-lived fission products have decayed. Also their accumulative fission yields are well known.

The procedure used to count the number of fissions was to tape a foil with the same composition as the samples to the outside of the transfer tube at the position adjacent to Godiva where samples were irradiated. After irradiation was completed, the foil was allowed to decay. The number of fissions in the sample was then found by counting the La-140 1,596 keV gamma activity relative to a Godiva calibration foil. After 13 days, the activity of La-140 is in transient equilibrium with, and therefore

proportional to, that of Ba-140 (half-life of 12.9 days). By comparing the La-140 1,596-keV gamma counts from the foil in question to these from a Godiva calibration foil, the number of fissions in the sample were calculated. The counting system consisted of a high-purity germanium detector with its associated electronics. The high-purity germanium detector was used to find the intensity of the 1,596 keV photopeak. Elaborate analysis of the 1,596-keV peak was unnecessary, since the peak was well separated from any other peak.

6. Discussion and Results

The parameters of the decay constants and group abundances for U-235 obtained in this experiment are listed in Table I, where in the last line the absolute yield is also reported and compared. Table I summarizes the fast neutron induced fission for U-235. The results listed were taken from separate nonlinear least squares fitting to all the data. As expected, infinite irradiation produced less error in the decay constants for groups 1 and 2, and instantaneous irradiation produced less error for groups 5 and 6. Groups 3 and 4 have larger error bars since neither type of irradiation techniques emphasize these groups.

Table I. Fast-fission delayed neutron data for U-235

Group #	**Decay Constant** $(sec)^{-1}$	**Relative Yield** (a_i/a)	**Absolute Yield** $(fissions)^{-1}$
1	0.0127 ± 0.0001	0.0395 ± 0.001	0.000644
2	0.0315 ± 0.0004	0.235 ± 0.005	0.00383
3	0.117 ± 0.0064	0.207 ± 0.008	0.00337
4	0.314 ± 0.0107	0.381 ± 0.011	0.00621
5	1.37 ± 0.0514	0.114 ± 0.005	0.00186
6	3.83 ± 0.1138	0.0235 ± 0.001	0.000383
Absolute Yield (neutrons/fission)			
This Experiment	Keepin et al.	Brady et al.	
0.0163 ± 0.0008	0.0165 ± 0.0005	0.0206 ± 0.002	

Note that a sound comparison of the data is only possible for the absolute yield, since the group abundances and decay constants are in some degree arbitrary (fitting six groups to the 271 known delayed neutron precursors). However, since the delayed neutron decay for U-235 and other fissionable isotopes can be satisfactorily fitted with six periods having in essence the same value, it can be inferred that six 'principal' precursors exist that dominate the decay, despite other perturbations. However, studies are under way to determine, if more than six dominant precursors exist. The absolute yield for U-235 (reported in Table I) determined by Brady et al. in 1989 has been recently reevaluated, and the new value calculated by Tal England is 0.0172.[16]

Table II presents the fast neutron-induced fission in Np-237 measured in this experiment. The delayed neutron data is also presented in the classical six-group representation established by Keepin. The last line in Table II compares the absolute yield value from this experiment to other available data.

Table II. Fast-fission delayed neutron data for Np-237.

Group	Decay Constant $(sec)^{-1}$	Relative Yield (a_i/a)	Absolute Yield $(fissions)^{-1}$
1	0.0123 ± 0.0009	0.032 ± 0.003	0.000403
2	0.0284 ± 0.0005	0.238 ± 0.006	0.00299
3	0.0971 ± 0.007	0.175 ± 0.008	0.00221
4	0.296 ± 0.014	0.360 ± 0.017	0.00454
5	0.914 ± 0.058	0.150 ± 0.014	0.00189
6	3.2 ± 0.13	0.045 ± 0.006	0.000567
Absolute Yield (neutrons/fission)			
This experiment		Malinkin et al.	Brady et al.
0.0126 ± 0.0007		0.0126 ± 0.0008	0.0114 ± 0.0012

The newly measured delayed neutron data for Np-237 compares very well to previously measured data in Dubna, Russia, but never published in English. This value[17] was given to us during a presentation of the results in the "Colloquium of Delayed Neutron Data" held in Obnisk, Russia, in April 1997.

Figure 3 shows the agreement between the experimental data and a curve calculated using the periods and abundances reported in Table I for instantaneous and infinite irradiations. Note that the graphed data only extends to the first 260 seconds; however, data was collected until the decaying delayed neutrons could not be distinguished from background neutrons.

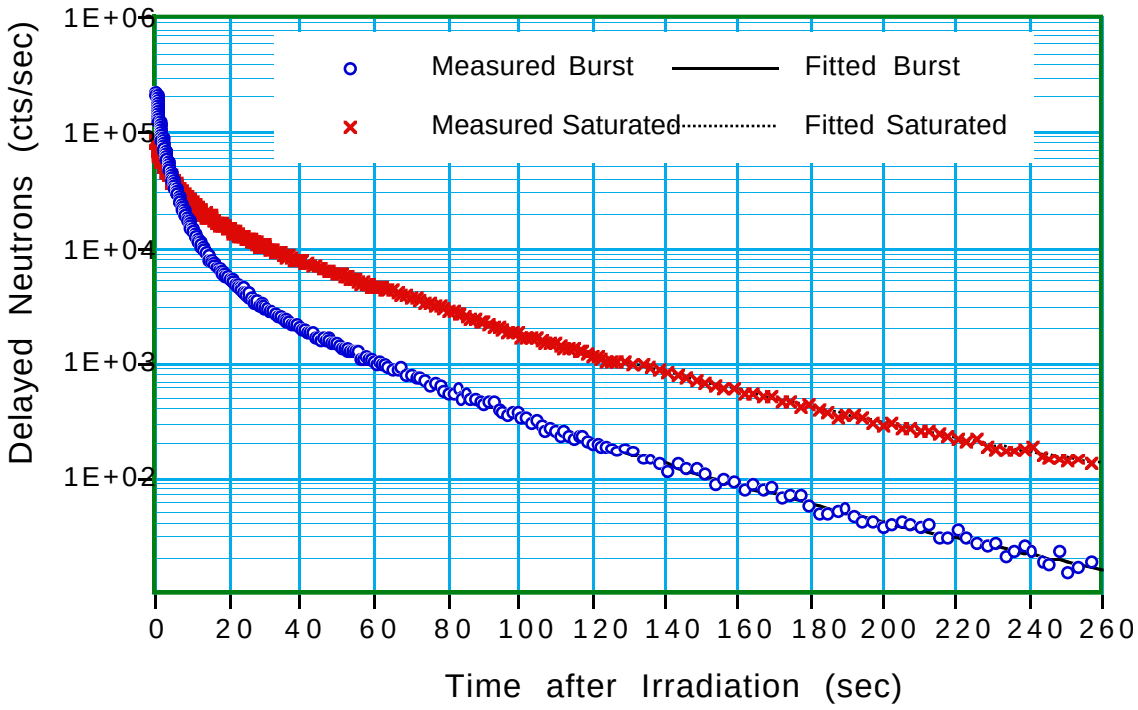

Figure 3. Measured delayed neutron decay data for saturated and burst irradiations shown with fitted data calculated from abundances and periods.

An appropriate check for the data in Table I can be made through the relation between reactivity and reactor period. This relationship is better known as the inhour equation. Figure 4 shows the inhour equation calculated using the parameters from this experiment and Godiva's results using Keepin's measurements. Keepin et al. performed measurements of period versus reactivity on the Godiva II assembly, which for this purpose was a geometrically clean assembly. The parameters measured in this experiment compare well with Keepin's measurements. Figure 4 provides one more check of the accuracy and validity of data presented in Table I.

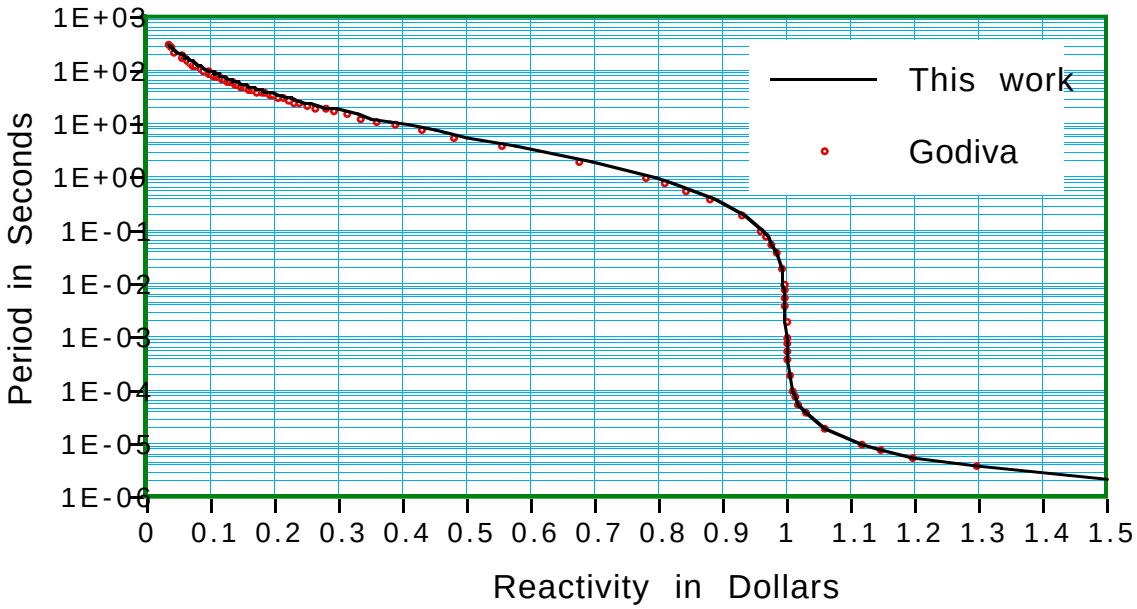

Figure 4. Inhour equation relationship for Godiva.

The period vs reactivity relationship used to plot Figure 4 is given by;

$$\rho(\$) = \frac{1}{\alpha \cdot T} + \sum_{i=1}^{6} \frac{a_i \frac{1}{\lambda_i}}{T + \frac{1}{\lambda_i}} \qquad (6)$$

where T is the reactor period in seconds, α is an experimentally measured value of 8.4 x 10^5 sec^{-1}, and a_i and λ_i are the relative abundance and decay constant, respectively, for the *i*th delayed neutron group.

The inhour equation results are also presented in tabular format in Table 3. The six-group representation from this experiment is compared to Keepin's parameters and to values from Brady and England to illustrate the differences. The time-dependent six-group representation of the delayed neutron emission from this experiment gives reactivities within 5% or less for periods smaller than 50 seconds when compared to the traditional Keepin values. The greatest difference is 6% for large periods. When this data is compared to Brady and England values, the greatest difference is 18% for long periods and about 11% near 0.4 $. Table III also demonstrates that the delayed neutron parameters from Brady and England tend to predict smaller reactivities in dollars for all periods compared to

this experiment and Keepin's data. Other researchers[3,4,5] have also noticed such a difference when using the delayed neutron parameters from Brady and England, which now are included in the ENDF/B-VI library.

Table III. Comparison of reactivity predictions from this experiment's parameters with recommended values for the Godiva assembly.

Period (sec)	Reactivity ($) using group constants from this experiment	Reactivity ($) using group constants from Brady's data.	Reactivity ($) using group constants from Keepin's data.	Relative difference between this experiment and Brady's data.	Relative difference between this experiment and Keepin's data
300	0.04062	0.03327	0.03811	0.181	0.0618
200	0.05809	0.04771	0.05452	0.177	0.0614
100	0.1027	0.08497	0.09653	0.173	0.0601
50	0.1697	0.1422	0.16000	0.162	0.0571
20	0.2935	0.2522	0.2788	0.141	0.0501
10	0.4085	0.3602	0.3912	0.118	0.0424
1	0.7944	0.7633	0.7818	0.0391	0.0157
0.6	0.8529	0.8297	0.8430	0.0272	0.0116
0.2	0.9360	0.9254	0.9312	0.0113	0.00513
0.1	0.9645	0.9588	0.9619	0.00591	0.00260
0.06	0.9776	0.9741	0.9760	0.00358	0.00163
0.02	0.9919	0.9909	0.9916	0.001008	0.000301
0.01	0.9958	0.9955	0.9958	0.000301	0.000010

7. Conclusion

The measurements reported in Table I for the decay constants and abundances are in substantial agreement with the well-established Keepin data reported nearly 40 years ago. The period reactivity relation arising from the measured yield was calculated using the parameters measured in this experiment and the six-group parameters of Keepin and Wimett. The results were the same within 1% for reactor periods equal or greater than one second. It is believed that the present calculation confirms and validates the data measured by Keepin and Wimett.

The newly measured Np-237 delayed neutron parameters have also been measured. The experimental uncertainties obtained in the measurement of the delayed neutron parameters are small. This is attributed to the high detector efficiency, sample size, and newly developed data acquisition system. The reported errors in the group parameters and the total yield values are mainly statistical. The good agreement between Keepin's data for U-235 and this experiment lends credibility to the analysis of the data and the experimental procedure.

In conclusion, the measured data for U-235 and Np-237 have been presented and compared to recent evaluations. In most cases the sets of experimental data compare well to each other. There are some differences when compared to computational methodologies. This experiment was able to measure the six-group delayed neutron parameters for Np-237. Therefore, it is recommended that the parameters from this experiment for Np-237 be included in new evaluations of the ENDF/B library. It is also recommended that new versions of the ENDF/B library place first priority to experimental data, and in the cases where experimental data lacks then theoretical data should be used. Based on this experiment there is no evidence for abandoning the use of the time-honored Keepin and Wimett data.

8. Acknowledgments

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