ABSTRACT

Recent interest in delayed neutron parameters including comparisons between macroscopic (experimental) and microscopic (calculated) results have prompted a set of experiments using the 1MW Triga Reactor at the Texas A&M University (TAMU) Nuclear Science Center (NSC) designed to measure the complete set of “seven-group” delayed neutron parameters for several higher actinides. Operating the Nuclear Science Center Reactor (NSCR) in a pulsed mode, a complete set of delayed neutron parameters were measured for Np-237 and Am-243. The total delayed neutron yield per 100 fissions for Np-237 and Am-243 was found to be 1.14 ± 0.07 and 0.85 ± 0.05, respectively. Comparisons to previous measurements are made where such measurements are available.

INTRODUCTION

In order to effectively operate a reactor with large loadings of higher actinide isotopes (e.g., an actinide burner reactor), accurate knowledge of each actinide isotope’s neutronic properties (including its delayed neutron yields and decay constants) is vital. The delayed neutron emission properties of the actinide isotopes (specifically U-235, Np-237, Am-241, and Am-243) have been actively studied at TAMU for several years. Due to a transfer time from core-to-detector of ~0.5 seconds, the previous experiments have been unable to measure the shortest-lived delayed neutron groups.

The existing experimental system has been modified to allow for pulsed experiments. One special feature of the NSCR is its ability to be operated in a pulsed mode. This feature allows the generation of extremely high neutron fluxes for very brief time periods. Thus, using the pulsing capabilities of the reactor, an irradiation could be performed that would accentuate the shorter-lived delayed neutron groups, allowing for a measurement of the complete “seven-group” delayed neutron emission parameters.

Previous work has shown that the use of a “seven-group” delayed neutron structure yields a superior fit to the measured data, has a more direct correlation to the precursor database, and has been shown to generate consistent decay constants independent of the fissioning isotope, compared to the traditional “six-group” structure suggested by
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KEEPING THE “SEVEN-GROUP” STRUCTURE WILL BE USED THROUGHOUT THIS WORK.

EXPERIMENTAL SETUP

The TAMU delayed neutron measurement system is composed of several integrated components (a pneumatic transfer system, a detector array, a storage container, and sensors) controlled by a computer with an internal I/O card. The sample to be irradiated is transferred to the core, to the detector array, and to a remote storage box via a pneumatic transfer system using CO₂ gas. A series of sensors transfer data to the computer relaying various information including sample position, permit control, and reactor pulse power. This section describes the individual components of this system; as well as how they interact with one another and the reactor console.

A. Computer Control System

The central portion of the delayed neutron measurement system is the computer control system (CCS). The CCS is designed to allow relatively hands-off execution of a particular delayed neutron measurement. This hands-off aspect of the system allows for simplicity in performing experiments, as well as increased repeatability of experiments by decreasing the potential for human error. The CCS interacts with the pneumatic system, controlling the position of the sample (receiver-core-detector-storage), timing the irradiation, and measuring the transfer time from core-to-detector. The CCS also reads the delayed neutron emissions versus time from the detector array. Lastly, the CCS inputs data from the reactor console. The reactor console data has two purposes: (1) to inform the experimenter of permit status and (2) to read the pulse power drawer for prompt burst irradiations. Thus, the computer is capable of determining when the reactor has been pulsed by reading the power from the reactor’s pulse power drawer. The computer can then immediately return the sample following a pulse. Figure 1 illustrates how the CCS interacts with the other components of the delayed neutron measurement system. The CCS functions are all performed through the use of a CIO-CTR board resident in the CCS computer.

The CCS uses a QBASIC program to perform all its actions. When executed, this program will prompt the user for all the necessary input data for a particular irradiation. The program will display the permit status and then wait to be prompted by the experimenter to begin the irradiation. The CCS will then transfer the sample into the core, allow it to irradiate until the reactor is pulsed, transfer the sample to the detector array (measuring the transfer time using a photosensor), collect the delayed neutron data versus time after irradiation, and lastly transfer the sample to a remote storage location.

B. Pneumatic Transfer System

The sample to be irradiated is transferred to and from the core using a pneumatic system (Fig. 2) consisting of pressurized CO₂ gas, 1” and 3/4” O.D. polyethylene tubing, a series of Swagelok Unions, and associated electronics and pneumatics. This pneumatic system connects all the individual components of the measurement system and provides a safe means of transporting the potentially radioactive samples to the detector array and to a remote storage location.
C. BF$_3$ Detector Array

The BF$_3$ detector array consists of three N. Wood Model G-20-5 BF$_3$ proportional counters embedded in a block of paraffin (Fig. 3). A thin lead sheath surrounds the sample tube to decrease the gamma-ray build-up in the detectors. Also, a thin cadmium sheet surrounds the detector array to eliminate any background sources of neutrons. The entire array is surrounded on all sides by 4" of lead to protect the surrounding personnel from any unnecessary doses.

The electronic components of the counting system are shown in Fig. 4. This system consists of a series of BF$_3$ detectors attached to preamps, high voltage bias, amplifiers, and single channel analyzers. The individual signals from the three detectors are combined using a dual-sum inverting amplifier. The signal then passes to the CCS computer that allocates the pulses to various time bins. A 50.8 nCi Cf-252 source has been used to calibrate the detector array. The energy response function for this array is shown in Fig. 5. Using this response function and the measured counts from the Cf-252 source, it was found that the detector efficiency for delayed neutrons was 2.17%. This efficiency has been confirmed using an AmLi neutron source borrowed from Los Alamos National Laboratory.

D. Actinide Samples

Two Np-237 and three Am-243 sources (fabricated by Isotopes Products Laboratory in California) were used during the irradiations. The samples consisted of an inner pellet 4.88 mm in diameter and 0.92 mm thick. The pellet was composed of a matrix of aluminum powder and actinide oxide. The aluminum and actinide were mixed together and pressed under high pressure to form the disc-shaped pellets. The pellets were encapsulated in a thin (0.05 mm) titanium cover.

The samples used were extremely pure. The Np-237 samples had an atomic purity of greater than 99.999% (assay date of March 3, 1983). The Am-243 samples had an atomic purity of greater than 99.987% (assay date of August 17, 1978). No attempt was made to correct for any impurities present in the samples. It should be noted that Am-243 decays to Pu-239, and this may cause some discrepancy in the Am-243 results when irradiated in a thermal neutron flux.

PROCEDURE

Several measurements have been performed to determine the short-lived delayed neutron group yields and half-lives for Np-237 and Am-243. Samples of each isotope were transferred into the NSCR via the quick pneumatic transfer system. The samples were irradiated in the B1 position at 300 W for approximately 10 seconds; then, the reactor was pulsed by adding $1.50$ reactivity. The pulses last for approximately 60 milliseconds. The CCS reads the power from the reactor console and removes the sample at the peak of the pulse. The sample is then transferred to the BF$_3$ detector array where the delayed neutrons versus time are counted. After counting, the samples are pneumatically transferred to a remote storage location. A high-purity germanium detector was used to determine the total number of fissions in the sample by measuring the buildup of certain fission products (specifically Ba-140, La-140, Ru-103, I-131, Mo-99, Zr-97, Sr-91, Te-132, I-132, and I-135). The
number of fissions were typically on the order of $10^9$. In the fission rate determination, the yield data for the fission products was taken from the ENDF/B-VI file.

RESULTS AND ANALYSIS

Parameters (yields and decay constants) for all seven groups of the alternate “seven-group” structure were acquired from the measured emission rates using a graphical stripping procedure. The delayed neutron curves were fit using the following relation:

$$C(t) = N_f \varepsilon \sum_i Y_i \lambda_i \exp(-\lambda_i t)$$

where $C(t)$ is the measured (dead-time corrected) delayed neutron count rates, $N_f$ is the measured total number of fissions during the irradiation, $\varepsilon$ is the detector efficiency, $Y_i$ is the delayed neutron yield for group $i$, $\lambda_i$ is the delayed neutron decay constant for group $i$, and $t$ is the time after the end-of-irradiation. The pulsing technique was unable to produce enough counts to allow for accurate measurement of the group 1 and 2a values. Thus, the results determined previously for group 1 were used to allow for a complete “seven-group” set. The group parameters determined in this work are presented in Table I.

For purposes of comparison, the total delayed neutron data from selected works from the literature are presented in Table II. As can be seen, the values measured here are in reasonable agreement with the few measured values available. The listed Np-237 results agree to within their associated standard deviations. The only results available for Am-243 are those calculated by Brady and England. These data were produced by a summation calculation using precursor data from ENDF/B-VI. However, the agreement between the total delayed neutron yields for Am-243 from Brady and England’s calculation and that measured here is excellent for such a high mass isotope.

These experiments illustrate the capability of the TAMU delayed neutron measurement system. The system is capable of measuring the complete set of delayed neutron parameters by coupling pulsing experiments (to yield groups 2b, 3, 4, 5, and 6) and finite length irradiations (to measure groups 1, 2a, 2b, 3, and 4). Calculations suggest that increasing the efficiency of the detector array may allow for measurement of all seven groups of the alternate “seven-group” structure using pulsing irradiations. This would yield the possibility of generating ultra-accurate relative yields by eliminating the need for the fission rate determination.

CONCLUSIONS

The experiments described here have allowed for complete measurement of the delayed neutron emission parameters for Np-237 and Am-243. The TAMU delayed neutron measurement system is currently capable of measuring delayed neutron emission parameters for all groups for nearly any nuclide. Experiments are planned to extend the above investigations to include other actinide isotopes (e.g., Pu-239, U-238, Am-241, etc.). Also, measurements will be performed for U-235 to allow for an accurate comparison of the TAMU system to the system used by Keepin. Investigations to increase the number of detectors (and hence the detector array efficiency) are undergoing. This modification is expected to allow
measurement of all groups of the "seven-group" structure in a single irradiation.

ACKNOWLEDGMENTS

The research presented here was funded under the auspices of the Japan/U.S. Actinide Program of Oak Ridge National Laboratory (ORNL) in cooperation with collaborators at the Japan Atomic Energy Research Institute (JAERI). The authors appreciate the interest in the development and improvement of nuclear data exhibited by T. Mukaiyama and H. Oigawa (JAERI). ORNL is operated by Lockheed Martin Energy Research Corporation under Contract No. DE-AC05-96OR 22464 with the U.S. Department of Energy.

REFERENCES


Detector Array

Pneumatic Transfer System

Control System

Reactor Console

Fig. 1. Schematic of the computer control system (CCS).

Bridge Level
(Elevation ~ 35 ft)

Chase Level
(Elevation ~ 25 ft)

Lower Research Level
(Elevation ~ 0 ft)

Sample Lines
Passive Divertors

Storage Box

Exhaust

Pneumatic Station

Gas Lines

Fig. 2. Pneumatic transfer system with approximate component altitudes indicated.
Fig. 3. BF3 detector array.

Fig. 4. BF3 detector array electronics.
TABLE I.
Delayed neutron yields and decay constants for preliminary pulsing experiments.

<table>
<thead>
<tr>
<th>Group</th>
<th>$\lambda_4$ (sec$^{-1}$)</th>
<th>$Y_i$ (neuts/100 fissions)</th>
<th>$\lambda_4$ (sec$^{-1}$)</th>
<th>$Y_i$ (neuts/100 fissions)</th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>0.0124 ± 0.0003</td>
<td>0.035 ± 0.002</td>
<td>0.0124 ± 0.0004</td>
<td>0.014 ± 0.004</td>
</tr>
<tr>
<td>2a</td>
<td>0.0283 ± 0.0007</td>
<td>0.183 ± 0.012</td>
<td>0.0283 ± 0.0009</td>
<td>0.192 ± 0.009</td>
</tr>
<tr>
<td>2b</td>
<td>0.0411 ± 0.0008</td>
<td>0.095 ± 0.005</td>
<td>0.0415 ± 0.0009</td>
<td>0.075 ± 0.005</td>
</tr>
<tr>
<td>3</td>
<td>0.155 ± 0.002</td>
<td>0.325 ± 0.018</td>
<td>0.151 ± 0.002</td>
<td>0.175 ± 0.009</td>
</tr>
<tr>
<td>4</td>
<td>0.397 ± 0.006</td>
<td>0.368 ± 0.015</td>
<td>0.392 ± 0.006</td>
<td>0.285 ± 0.009</td>
</tr>
<tr>
<td>5</td>
<td>0.845 ± 0.03</td>
<td>0.100 ± 0.011</td>
<td>0.895 ± 0.04</td>
<td>0.075 ± 0.008</td>
</tr>
<tr>
<td>6</td>
<td>2.55 ± 0.03</td>
<td>0.037 ± 0.009</td>
<td>2.45 ± 0.04</td>
<td>0.035 ± 0.007</td>
</tr>
<tr>
<td>total:</td>
<td>1.14 ± 0.07</td>
<td></td>
<td>total:</td>
<td>0.85 ± 0.05</td>
</tr>
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TABLE II.
Total delayed neutron yields for Np-237 and Am-243 as compared to values from the literature.

<table>
<thead>
<tr>
<th>$\nu_b$ (delayed neutrons per 100 fissions)</th>
<th>Np-237</th>
<th>Am-243</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waldo et al. 7</td>
<td>1.068 ± 0.098</td>
<td>-</td>
</tr>
<tr>
<td>Benedetti et al. 8</td>
<td>1.22 ± 0.03</td>
<td>-</td>
</tr>
<tr>
<td>Brady and England 9</td>
<td>1.14 ± 0.12</td>
<td>0.80 ± 0.09</td>
</tr>
<tr>
<td>This work</td>
<td>1.14 ± 0.07</td>
<td>0.85 ± 0.05</td>
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