Title: NEUTRON ENERGY MEASUREMENTS FOR IMPURE PLUTONIUM SAMPLES

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Neutron Energy Measurements for Impure Plutonium Samples

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I. ABSTRACT

This report describes a technique used to measure the average neutron energy on impure plutonium samples. This measurement is done using the Epithermal Neutron Multiplicity Counter (ENMC). The measurement of neutron energy is done at the same time as the accountability or safeguards measurement. Neutron totals and coincidence measurements of plutonium have the problem that impurities present in sample add to the observed neutron counts because of alpha particle induced source neutrons and the related multiplication. The primary signal that is needed to measure the \(^{240}\text{Pu}\) effective mass is the spontaneous fission rate; however, induced fissions related to the impurities increase the observed response. In most cases, the \((a,n)\) source neutrons have an energy distribution that is different than for spontaneous fission. Thus, the ability to measure the neutron energy distribution helps in the identification of impure plutonium samples. A description of the ENMC detector components and discussion on the average neutron energy calibration are provided.

II. INTRODUCTION

Neutron detectors using \(^3\text{He}\) tubes have been used extensively for quantitative measurements of plutonium for safeguards of nuclear material and nuclear material control and accountability.\(^1\) Passive measurements are made using neutron coincidence counting or multiplicity counting to obtain the \(^{240}\text{Pu}\) effective mass. This is a direct measurement of the even numbered plutonium isotopes present in the item being measured. The total plutonium mass is determined by using the \(^{240}\text{Pu}\) effective mass and the plutonium isotopic ratios for the item being measured. Neutron totals and coincidence measurements of plutonium can be biased by impurities that may be present in the item being measured as impurities can add to the observed neutron counts. The mode in which this happens most commonly is the alpha particle induced source neutrons \((a,n)\) and the related multiplication. Multiplication is the degree to which a sample or item induce neutrons from either spontaneous fission or \((a,n)\) reactions.\(^2\) Since the primary signal needed to measure the \(^{240}\text{Pu}\) effective mass is the spontaneous fission rate, any induced fissions related to impurities present increases the observed response in the neutron totals and coincidence measurements. In most cases, the \((a,n)\) source neutrons have an energy distribution that is different than for spontaneous fission neutrons. Thus, the ability to measure the neutron energy distribution can help in the identification of impure plutonium samples.

We have extended the capability of the high efficiency Epithermal Neutron Multiplicity Counter (ENMC)\(^3\) to include the measurement of the average neutron energy. The ENMC is a neutron well counter that shares some basic design features with other neutron well counters. One of these design features is the arrangement of the \(^3\text{He}\) detector tubes in concentric rings. The ENMC design has four rings of detector tubes, with the innermost ring called Ring 1 and the outmost ring called Ring 4. The average neutron energy measurement is done concurrently with the standard multiplicity measurement used to determine the \(^{240}\text{Pu}\) effective mass, using the
totals rates from the innermost and outmost rings of detectors. The ratio of the totals of Ring 4 to Ring 1 provides good sensitivity to determine the average neutron energy of the item being measured. The system was calibrated for this neutron energy response using neutron sources with a wide range of energy including AmLi (~ 0.3 MeV), Pu metal (~ 2 MeV), PuF4 (~ 1.3 MeV), 252Cf (~ 2.14 MeV), and PuB (~ 2.8 MeV). A PuBe neutron source was also used for the energy calibration but its measured energy was significantly below the published average energy of ~ 4.2 MeV. This energy reduction was probably the result of secondary neutron reactions in the Be in addition to the (a,n) reactions.

With this added capability, data from the ENMC can be studied and changes in the composition of the items that have been measured can be detected and identified. Having this knowledge, adds another piece of information that can strengthen an inspector’s ability to authenticate instrument’s operational status as well as provide information on the facility activities. This can be done without making any changes to the measurement routine including the measurement time. The paper presents the ENMC detector tube tests and calibration, with the added capability for neutron energy measurements.

III. DETECTOR DESCRIPTION

A. Helium-3 Tubes

The ENMC is differentiated from other neutron well counters by its high-efficiency and short-die-away time. These characteristics are the result of careful moderator design and the use of the 3He detector tubes with 10-atm pressure and proprietary gas mix to facilitate fast-pulse collection time. The development of the detector tube was done by Reuter-Stokes. In general, the 3He tubes have a slow ionization charge collection time, and special gas additives are required to give a fast charge collection that is compatible with the use of AMPTEK amplifiers. If the pulse charge collection is too slow, the short time constants in the AMPTEK amplifiers (190 ns) will give double counting for the larger pulses, resulting in disproportionate counting rates for the measured parameters; Singles, Doubles, and Triples that are used in the multiplicity analysis. This concern is minimized with the development of the Reuter-Stokes 10-atm detector tubes and provides the basis on which the ENMC is built.

The ENMC uses Reuter-Stokes tubes with the specifications shown in Table 1.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of tubes @ 28 in. (RS-P4-0828-105)</td>
<td>121</td>
</tr>
<tr>
<td>Diameter</td>
<td>25.4 mm</td>
</tr>
<tr>
<td>Active length</td>
<td>711 mm</td>
</tr>
<tr>
<td>He pressure</td>
<td>10 atm</td>
</tr>
<tr>
<td>Cathode</td>
<td>Al</td>
</tr>
<tr>
<td>Operating bias</td>
<td>1720 V</td>
</tr>
<tr>
<td>AMPTEK (A111) amplifiers</td>
<td>27</td>
</tr>
</tbody>
</table>
B. Moderator and Electronics

A diagram of the ENMC is shown in Figure 1. The sample cavity is 193.6 mm (7.62") in diameter and 430 mm (16.93") tall.

![Diagram of the ENMC showing the layout of the 3He detector tubes in HDPE detector moderator.](image)

**Figure 1.** Diagram of the ENMC showing the layout of the 3He detector tubes in HDPE detector moderator.

C. Multiplicity Electronics

The ENMC can operate in both the standard neutron coincidence mode as well as the multiplicity mode. In both modes, the primary source of statistical uncertainty is the pileup of accidental counts in the coincidence gate. By pairing the ENMC with the Advanced Multiplicity Shift Register (AMSR) the accidental error is reduced by about a factor of 1.4.\(^5\) The AMSR provides for fast accidental sampling. The fast accidentals sampling feature incorporated in the AMSR samples the accidentals gate at the clock speed of the computer board on the shift register leading the improvement of the statistical error on the accidentals determination.

To reduce the counter's dead time, there are 27 AMPTEK A111 amplifiers to reduce the pulse rate through each amplifier. The signals are input to a de-randomizer board before input to the AMSR.

The AMSR is shown in Figure 2 and the auxiliary input connections on the back of the AMSR are shown in Figure 3.
The AMSR-150 Shift register includes High Voltage (HV) and low voltage power supplies and used for neutron multiplicity counting.

Figure 3. Back panel on the AMSR-150 shift register which includes two auxiliary scalar inputs.

The detector connectors are shown in Figure 4 and include the four separate detector rings as well as the sum of all of the rings. The singles from Ring 1, which contains 21 detector tubes, are routed into the auxiliary scalar Number 1 of the Advanced Multiplicity Shift register (AMSR), and the Singles from Ring 4, which contains 40 detector tubes, are routed auxiliary scalar Number 2. The auxiliary inputs on the AMSR are limited to only measuring the singles rates. The full detector system is shown in Figure 5.

Figure 4. Detector signal outputs on the HV distribution box for the ENMC. The outputs that are used in the present application are ring 1 (to Aux. 1), ring 4 (to Aux. 2), and the sum of the four rings (to signal in of AMSR).
IV. AVERAGE NEUTRON ENERGY CALIBRATION

A. High Voltage Plateau

The detector bias plateau, also called the high voltage plateau, is shown in Figure 6. The data presented in Figure 6 shows the normalized data for a single amplifier compared to the data from the sum of the 27 amplifiers. The comparison made in Figure 6 reveals that for a typical representative channel the gain in each of the amplifiers have been matched so that the plateau from a single amplifier have the same operating characteristics as does the system as a whole.
B. Ring Ratios for Average Neutron Energy

A series of measurements were made using $^{252}$Cf, AmLi, and plutonium with impurities to characterize the ENMC sensitivity to detecting differences in neutron energy. The ratio of the singles measured in Ring 4 to those measured in Ring 1 for these neutron sources is given in Table 2 and shown in Figure 7. The ring ratio method provides an approximation for the average energy for a sample of unknown plutonium mass and composition measured in the ENMC. This energy information can provide additional information on the impure plutonium samples.

Table 2. Average neutron energy for material measured and ring ratio results.

<table>
<thead>
<tr>
<th>Material Type</th>
<th>Average Neutron Energy</th>
<th>Ring ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>AmLi</td>
<td>0.3</td>
<td>0.26</td>
</tr>
<tr>
<td>PuF$_4$</td>
<td>1.3</td>
<td>0.50</td>
</tr>
<tr>
<td>PuO$_2$</td>
<td>2</td>
<td>0.59</td>
</tr>
<tr>
<td>PuO$_2$</td>
<td>2</td>
<td>0.60</td>
</tr>
<tr>
<td>$^{252}$Cf</td>
<td>2.14</td>
<td>0.59</td>
</tr>
<tr>
<td>PuB</td>
<td>2.9</td>
<td>0.78</td>
</tr>
</tbody>
</table>

Figure 7. The counting rate ratio for neutron sources of different energy.
V. SUMMARY

The primary application of the ENMC is to measure bulk samples of dirty scrap plutonium using multiplicity counting.

The ENMC has excellent performance characteristics. The ENMC detector ring ratios can be used to determine the average energy of the sample neutrons. The large separation between the inner and outer rings, as illustrated in Figure 1, make it possible to have measurable energy dependence captured in the ring ratio. The (α, n) neutrons from plutonium contaminated with fluorine have a lower energy than the spontaneous fission neutrons from $^{240}$Pu effective, and this type of contamination can be identified with the system. This is notable because fluorine is one of the most common contaminants making assay difficult.

VI. ACKNOWLEDGMENT

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VII. REFERENCES


