NEUTRON MULTIPLICITY DRUM MONITOR

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ABSTRACT

The potential for improved measurement accuracy and the ability to gain sample matrix information as part of a waste assay are but two of the advantages of multiplicity measurements of waste. Neutron Multiplicity Counting (or Neutron Time Correlation Analysis) using the point source interpretation model allows one to determine three of the four unknown assay parameters by measuring the Totals count rate, the Reals count rate (correlated pairs) and the correlated Triples rate. Thus, where one of the unknown assay parameters can be established or is known, then absolute measurements of waste are possible.

The current collaborative development of a multiplicity drum monitor for the measurement of plutonium contained in 200 litre waste drums is described in the present paper. Although the device may be employed with conventional shift register based coincidence electronics for pair correlation analysis, it has been designed specifically for neutron multiplicity counting using the Time Correlation Analyzer (TCA) for triple correlation analysis.

Data is presented for measurements on a prototype drum monitor developed at the Joint Research Centre. The recent evolution of the design, both in order to optimize measurement performance and to satisfy specific customer requirements, is described in this paper.

INTRODUCTION

Plutonium present in radioactive waste, generates neutrons from spontaneous fission events, from \((\alpha,n)\) reactions and induced fission events caused by primary neutrons. Passive neutron multiplicity counting (neutron triple correlation) provides a means of determining three of the four unknown parameters established in the point source interpretation model. These unknown parameters include the spontaneous fission rate \(F_s\) of the plutonium in the sample, the sample \((\alpha,n)\) reaction rate, the sample multiplication (induced fission rate) and the detector (including sample) detection probability or detection efficiency. The spontaneous fission rate \(F_s\) is always treated as an unknown as it is used to determine the \(Pu\ 240\) effective mass (MPu240) of the plutonium and hence the total plutonium content of the sample.

Using multiplicity counting, three quantities may be derived from experimental data using the Time Correlation Analyzer (TCA). These three quantities are the effective numbers of singles \(R_1\), the correlated doubles (pairs) \(R_2\) and the correlated triples \(R_3\). Note that \(R_1\) is the Totals count rate and \(R_2\) is the Reals count rate (R) familiar in conventional coincidence counting. These measured
quantities are related to physically useful quantities (known and unknown parameters of the point source model) by the following three equations:

\[ R_1 = \varepsilon F_S T_M M v_{s(1)} (1 + \alpha) \]

\[ R_2 = \varepsilon^2 F_S T_M M^2 \left[ v_{s(2)} + (M-1) \frac{v_{s(1)} v_{i(2)} (1 + \alpha)}{v_{i(1)}^{-1}} \right] \]

\[ R_3 = \varepsilon^3 F_S T_M M^3 \left\{ v_{s(3)} + 2(M-1) \frac{v_{s(2)} v_{i(2)} + (1 + \alpha) (M - 1) \frac{v_{s(1)} v_{i(3)} + v_{i(1)}^{-1}}{v_{i(1)}^{-1}}}{2(M-1) v_{i(2)}^{-1}} \right\} \]

In these equations the symbols have the following meaning:

- \( S_{\alpha} = (\alpha, n) \) neutron reaction rate of the sample
- \( \varepsilon \) = probability for detection of a neutron
- \( F_S \) = spontaneous fission rate of the sample
- \( M \) = fast neutron multiplication factor

\[ \alpha = \frac{S_{\alpha}}{v_{s(1)} F_S} \]

The detailed characteristics of the 3rd Generation Neutron TCA have been described elsewhere (Ma93a, Ma93b). Unlike other multiplicity counting instruments which use a single gate, the TCA employs 16 simultaneous gates (or observation intervals) of differing lengths. It uses three trigger methods for data acquisition and each trigger method opens 16 simultaneous gates. In addition to the conventional signal trigger method, the TCA includes the periodic trigger method and the delayed signal trigger method. This latter trigger method provides a set of 16 gates to accumulate the accidental counts.

The TCA allows a variety of measurement and analysis methods based on the point source interpretation model (Ma95). The analysis methods which can be employed with the drum monitor are summarized below:

1. Total neutron counting (1 equation in \( R_1 \))

Total neutron counting is used in order to reach the lowest detection limits. All of the neutron signals are assumed to arise from the spontaneous fission of Pu. In this mode of operation the internal Cd liner of the drum monitor may be removed to increase the detection probability.

2. Pair Correlation (2 equations in \( R_1 \) and \( R_2 \))

2.1 Routine FE

Known \( \alpha \) ratio and multiplication \( M \)

Unknown \( F_S, \varepsilon \)

2.2 Routine FM

Known \( \alpha \) ratio and detection probability \( \varepsilon \)

Unknown \( F_S, M \)

In this method a two parameter analysis is performed so that the spontaneous fission rate and one other unknown parameter may be determined. The values of the two other unknown parameters from the point source interpretation model must be established by other means or be known.
3. Triple Correlation (3 equations in $R_1$, $R_2$, and $R_3$)

3.1 Routine FEM
- Known $\alpha$ ratio
- Unknown $F_S$, $e$, $M$

3.2 Routine FESA
- Known Multiplication (or $M = 1$)
- Unknown $F_S$, $e$, $S_\alpha$

3.3 Routine SAFM
- Known Detection probability $e$
- Unknown $S_\alpha$, $F_S$, $M$

In this method a three parameter analysis is performed so that the spontaneous fission rate and two other unknown parameter may be determined. The value of the other unknown parameter must be established by other means or be known.

4. Pair Correlation counting using calibration sources.

This is equivalent to conventional coincidence counting where calibration standards covering a range of Pu masses and with an equivalent matrix to that of the sample are used to establish a calibration curve. The Pu 240 effective mass (MPu240) is plotted as a function of the correlated pairs rate $R_2$ (or the Reals count rate). Once the Reals rate $R_2$ is measured for a given sample the MPu240 may be determined from the calibration curve.

**CHARACTERISTICS OF THE JRC PROTOTYPE MULTIPICITY DRUM MONITOR**

The drum monitor is a relatively high efficiency $4\pi$ detector with He-3 detector tubes arranged in a decagon. With the internal cadmium liner fitted the maximum detection probability is between 15 % and 20 %. The internal cadmium liner may be removed easily for higher efficiency Totals counting.

Typical neutron decay time for the JRC prototype drum monitor is about 70 $\mu$ sec. using a total of 64 He-3 detector tubes. Neutron decay time data is presented in Fig. 1. It can be seen in Fig. 1. that, with the internal Cd liner fitted, the detector exhibits a single exponential decay for a period of over 300 $\mu$ sec. It is an important requirement for the application of the point source interpretation model that the detector have a single exponential decay constant.

Several of the characteristics of the multiplicity drum monitor may be varied in order to change the performance of the device. The internal Cd liner may be removed easily for higher efficiency Totals counting. The detector modules may be replaced with modules containing more He-3 detector tubes or detector tubes with different characteristics. The internal polyethylene shielding may be increased.

**MEASUREMENT RESULTS**

Measurements of the axial variation of the maximum detection probability for the JRC prototype detector using a small Cf-252 source are tabulated in Table 1, with no matrix present in the measurement chamber. The data were obtained using an earlier prototype TCA which used only the signal trigger method. For measurements on the central axis, the Totals, Reals and correlated Triples are tabulated with values of the detection probability (maximum detector efficiency) determined using the analysis methods FE (pair correlation) and FEM and FESA (triple correlation). In the final column the variation of the detection probability is tabulated for measurements made at the periphery of the chamber, close to the He-3 detector modules.

In the analysis the values of the unknown parameters have been provided and the point source model has been used to determine the detection probability from the measured neutron pulse train data. From the results it can be seen that the detection probability varies as one would expect with the maximum values at the bottom of the chamber for the central measurements and the maximum values in the middle of the chamber for the peripheral measurements.
<table>
<thead>
<tr>
<th>Sample Position (cm)</th>
<th>R1 Totals (sec(^{-1}))</th>
<th>R2 Reals (sec(^{-1}))</th>
<th>R3 Correlated Triples (sec(^{-1}))</th>
<th>Pair Correl. FE</th>
<th>Triple Correl. FEM</th>
<th>Triple Correl. FESA</th>
<th>Detection Probability Epsilon (%)</th>
<th>Detection Probability Epsilon (%)</th>
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</thead>
<tbody>
<tr>
<td>0</td>
<td>2796.5</td>
<td>758.68</td>
<td>113.79</td>
<td>17.062</td>
<td>17.116</td>
<td>16.992</td>
<td>12.686</td>
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<td>662.78</td>
<td>94.16</td>
<td>15.920</td>
<td>15.785</td>
<td>16.096</td>
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<td>92.34</td>
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<td>15.978</td>
<td>15.809</td>
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<td>15.858</td>
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<td>625.96</td>
<td>86.41</td>
<td>15.482</td>
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<td>14.847</td>
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Table 2  Pu Sample No. 22 (MPu240=2.738g) in Central Position of Empty Cavity.

<table>
<thead>
<tr>
<th>Sample Position (cm)</th>
<th>R1 Totals (sec(^{-1}))</th>
<th>R2 Reals (sec(^{-1}))</th>
<th>R3 Corr. Triples (sec(^{-1}))</th>
<th>MPu240 (g)</th>
<th>Epsilon (%)</th>
<th>MPu240 (g)</th>
<th>M</th>
<th>Epsilon (%)</th>
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</thead>
<tbody>
<tr>
<td>0</td>
<td>824.49</td>
<td>94.61</td>
<td>11.876</td>
<td>2.768</td>
<td>16.867</td>
<td>2.787</td>
<td>1.0573</td>
<td>16.722</td>
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<td>10</td>
<td>776.88</td>
<td>84.29</td>
<td>10.051</td>
<td>2.759</td>
<td>15.948</td>
<td>2.789</td>
<td>1.0588</td>
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<td>25</td>
<td>770.48</td>
<td>82.99</td>
<td>9.739</td>
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<td>744.86</td>
<td>78.09</td>
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<td>2.703</td>
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<tr>
<td>85</td>
<td>711.21</td>
<td>71.10</td>
<td>7.769</td>
<td>2.741</td>
<td>14.695</td>
<td>2.757</td>
<td>1.0570</td>
<td>14.585</td>
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</table>

Table 3  Pu Sample No. 22 (MPu240=2.738g) in Central Position of Concrete Drum.

<table>
<thead>
<tr>
<th>Sample Position (cm)</th>
<th>R1 Totals (sec(^{-1}))</th>
<th>R2 Reals (sec(^{-1}))</th>
<th>R3 Corr. Triples (sec(^{-1}))</th>
<th>MPu240 (g)</th>
<th>Epsilon (%)</th>
<th>MPu240 (g)</th>
<th>M</th>
<th>Epsilon (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>628.68</td>
<td>54.719</td>
<td>5.0564</td>
<td>2.814</td>
<td>13.055</td>
<td>2.732</td>
<td>1.0486</td>
<td>13.113</td>
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<tr>
<td>20.5</td>
<td>313.40</td>
<td>12.077</td>
<td>0.434</td>
<td>3.134</td>
<td>5.664</td>
<td>2.813</td>
<td>1.0196</td>
<td>5.529</td>
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<tr>
<td>41.0</td>
<td>260.96</td>
<td>8.166</td>
<td>0.269</td>
<td>3.213</td>
<td>4.599</td>
<td>3.13</td>
<td>1.0461</td>
<td>4.764</td>
</tr>
<tr>
<td>61.5</td>
<td>366.13</td>
<td>16.748</td>
<td>0.786</td>
<td>3.084</td>
<td>6.724</td>
<td>2.94</td>
<td>1.0389</td>
<td>7.161</td>
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<tr>
<td>82.0</td>
<td>566.70</td>
<td>44.827</td>
<td>3.912</td>
<td>2.760</td>
<td>11.627</td>
<td>2.78</td>
<td>1.06</td>
<td>11.408</td>
</tr>
</tbody>
</table>
As a part of the evaluation of the multiplicity drum monitor, preliminary measurements have been made using a well characterized plutonium sample with a known Pu 240 effective mass (MPu240) of 2.738 g. Measurements have been made with the sample at different positions on the central axis of the drum. In Table 2 data are presented for both pair correlation (method FE) and triple correlation (method FEM) for measurements with no matrix present.

In order to obtain the two parameter pair correlation result the known values of self multiplication \( M \) and the alpha ratio have been supplied to the analysis algorithm which has been used to calculate both MPu240 and the detection probability \( \text{Epsilon} \). Very good agreement with the declared value of MPu240 has been achieved and the triple correlation FEM analysis has correctly determined the multiplication, \( M \).

In Table 3 data are presented for equivalent measurements with a concrete drum matrix. Again, very good agreement with the declared value of MPu240 is achieved despite a difficult matrix. The pair correlation result is better than one might expect as the algorithm has been supplied with the correct values of the unknown parameters \( M \) and the alpha ratio.

**DRUM MONITOR MODIFICATIONS**

As a result of experience using the JRC prototype multiplicity drum monitor and in response to specific user requirements, a number of modifications are being made to the design. These modifications involve both optimization of design parameters and changes to the drum loading mechanism and detector electronics.

The modified multiplicity drum monitor will employ a drum carrier moving on rails in place of the hoist loading system of the JRC prototype. This modification simplifies some aspects of the design and facilitates a greater degree of automation in the drum loading process. A manually operated drum lifter is used to place the drum on the carrier.

In order to reduce electronic noise, sealed low profile preamplifier-discriminator junction boxes are being implemented employing the AmpTek A-III preamplifier-discriminator. The concept for the new junction box was inspired by earlier work at Los Alamos (SI91). In the new design He-3 detector tubes can be changed without dismantling the junction box and a desiccant is not required. The sealed high voltage region of each junction box can be dismantled for maintenance should this be required.

![Image](image_url)

**Fig. 1 Decay Time Data**

The preamplifier-discriminator output from each junction box will be connected to a de-randomizing buffer-mixer-counter circuit implemented using VLSI technology. The mixer circuit will improve the counting statistics for pair and triple correlation and the device has independent counters on each input channel that provide a useful diagnostic feature. The counters can also be used to assess azimuthal variations of the distribution of neutron sources in a drum.

Finally, the dimensions of the measurement chamber are being optimized in keeping with specific user requirements. These modifications will have the effect of improving
the axial linearity and increasing the maximum value of the detection probability, as well as reducing the background count rate.

CONCLUSION

The multiplicity drum monitor has advantages over conventional neutron coincidence counting for a variety of waste types as demonstrated by the measurement of simulated waste samples. For samples where either the multiplication $M$ or the alpha ratio are known or can be determined, the use of the multiplicity drum monitor constitutes an absolute measurement method.

Measurements of background counting rates using the JRC prototype have demonstrated that, provided adequate counting statistics can be obtained, accurate measurements of very small Pu samples (<100 mg) may be made with the multiplicity drum monitor. The modifications to the design will improve the measurement performance for specific waste drum types by reducing electronic noise, by enhancing the detection efficiency and by facilitating alternate drum loading methods.

REFERENCES


