THE ASSAY OF Pu BY NEUTRON MULTIPlicity COUNTING USING PERIODIC AND SIGNAL TRIGGER METHODS

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ABSTRACT

For the assay of Plutonium neutron multiplicity counting is used with both periodic and signal triggered multiple observation intervals. The applied triple correlation method has the advantage of providing three independent equations for each method and interval. In addition to the spontaneous fission rate ($F_S$), one can solve for two other unknowns from the multiplication ($M$), the detection probability ($\epsilon$) and the ($\alpha$,n) reaction rate ($S_{\alpha}$). In most practical cases of bulk material and waste one or more of these parameters are well defined and the method constitutes an absolute measurement technique. Measurement results have indicated particular advantages for each observation interval trigger method. Bulk material is easily measured with the signal trigger method. In contrast, the periodic trigger method is consistently better for material with a small Pu mass and a high alpha ratio. This can be understood by comparing the three equations of the factorial moments of the measured neutron multiplicity distributions from the two interval trigger methods. Averaging the assay results over several observation intervals reduces the error of the result for both trigger methods compared with a single time channel multiplicity counter. Consistency between the counts of different observation intervals improves confidence in the measurement result. These conclusions are illustrated with experimental data.

INTRODUCTION

Neutrons from spontaneous fission events, from (\alpha,n) reactions and from induced fission events are generated by plutonium, whether it is present in radioactive waste or in bulk form. As a relative technique, neutron pair correlation (Bi74) using the shift register based coincidence counter (Bo75, En79) permits the determination of the spontaneous fission rate $F_S$ of the plutonium. If the isotopic composition of the Pu in the sample is known then the ratio formed by the ($\alpha$, n) reaction rate and the spontaneous fission neutron emission rate $\nu_{\alpha,n} F_S$ can be calculated. If neutron multiplication is neglected, then the detection probability $\epsilon$, which depends on the self shielding of the Pu in the matrix, can be eliminated. In this way $F_S$ and hence the plutonium mass may be determined from the two experimental quantities derived from the pair correlation measurement, the effective number of neutron singlets $R_1$, and the effective number of correlated neutron doublets $R_2$. The quantity $R_2$ is more commonly known as the Reals count, $R$ when derived from a coincidence counter.

In contrast with coincidence counting (pair correlation) neutron multiple correlation
(neutron multiplicity counting) is an absolute technique. It is an extension of neutron coincidence counting and considers singlet neutrons, correlated doublets (pair correlation) and in addition triple correlated neutrons. The technique is particularly applicable if samples are not well characterized or when the calibration is inappropriate. In this situation large errors result for coincidence counting and no indication of the failure of the pair correlation technique is provided. By providing data which permits the solution of three of the four unknowns \((F_s, S_\alpha, \epsilon \text{ and } M)\) multiplicity counting is a more robust measurement method for the determination of plutonium mass.

Both pair and triple neutron correlation analysis can be carried out using the neutron Time Correlation Analyser (TCA) which will be described later. This paper discusses the advantages of the use of neutron multiple correlation analysis as implemented using the 3rd generation neutron Time Correlation Analyser for the measurement of bulk material and waste containing plutonium. Results are presented from measurements of real waste using a prototype TCA.

**INTERPRETATION MODEL FOR PLUTONIUM MASS DETERMINATION**

An interpretation model for the determination of plutonium mass based on the measurement of the spontaneous fission rate using triple neutron correlation has been developed by Hage (Ha85, Ha86, Ci86). With the triple neutron correlation technique using the TCA, three quantities may be derived from experimental data. These are the the effective numbers of singlets \(R_1\), correlated doublets \(R_2\) and correlated triplets \(R_3\). These measured quantities are related to physically useful quantities by the following three equations:

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

\[
R_2 = \epsilon^2 F_s T_M M^2 \left[\frac{v_{s(2)} + (M-1)}{v_{I(1)}-1}\right] v_{I(1)} v_{I(2)}(1+\alpha)
\]

\[
R_3 = \epsilon^3 F_s T_M M^3 \left\{\frac{v_{s(3)} + 2(M-1)}{v_{I(1)}-1} \frac{v_{s(2)} v_{I(2)}}{v_{I(1)}-1} (1+\alpha) (M - 1)\right\}
\]

\[
\frac{v_{s(1)}}{v_{I(1)}-1} \left[\frac{v_{I(3)} + 2(M-1)}{v_{I(1)}-1} \frac{v_{I(2)}}{v_{I(1)}-1}\right]
\]

In these equations the symbols have the following meaning:

- \(S_\alpha = (\alpha,n)\) neutron reaction rate of the sample
- \(M = \) fast neutron multiplication factor
- \(\epsilon = \) probability for detection of a neutron

\(F_s = \) spontaneous fission rate of the sample

\[\alpha = \frac{S_\alpha}{v_{s(1)} F_s}\]
The implementation of the interpretation model in software and its use in plutonium mass determination is described in the next section in conjunction with the description of the TCA.

TIME CORRELATION ANALYSER

The development of the 3rd generation neutron Time Correlation Analyser (TCA) is based on years of experience at JRC Ispra (B174, B079). The instrument combines the features of the coincidence counter (for neutron pair correlation) and multiplicity counter (for neutron triple correlation). It incorporates features for the measurement of both plutonium bearing waste and bulk plutonium.

Essentially the TCA is an instrument for determining the numbers of signal groups or signal multiples which arrive from the detector head and which correspond to neutrons detected in the detector system. In contrast to the shift register coincidence counter and other multiplicity counters which operate using only the signal trigger method, the TCA operates using two independent measurement methods, the signal trigger method and the random or periodic trigger method. In the latter method each observation interval is opened periodically by an external trigger, without any delay from interval to interval. The observation interval or gate width is not triggered by the detection of an initial neutron.

For both the signal and periodic trigger measurement methods the TCA obtains a result for each of 16 simultaneously triggered observation intervals (gate widths) of duration $\tau_0$, 2 $\tau_0$, ..., 16 $\tau_0$. Aside from facilitating the determination of detector decay constant and dead time, reduced error assay results are obtained by averaging the results corresponding to the observation intervals spanning the decay time of the detector. The variation of the assay result between different observation intervals is also a useful measurement diagnostic.

The use of the TCA permits the solution of three equations for the determination of three unknowns. In addition to $F_S$ (the spontaneous fission rate), it is possible to solve for two unknowns selected from the three parameters $S_\alpha$ (the ($\alpha$,n) reaction rate), $\epsilon$ (the detection probability of the unknown Pu distribution in the sample) and $M$ (the neutron multiplication).

For example, if $S_\alpha$ is known (because the isotopic composition is known) then one can determine $\epsilon$, $M$ and $F_S$. If $\epsilon$ is known then one can determine $S_\alpha$, $M$ and $F_S$. Finally if $M$ can be eliminated then $\epsilon$, $S_\alpha$ and $F_S$ can be determined. The last two cases are of importance where the sample isotopic ratio is not known. In many situations either $\epsilon$, $S_\alpha$ or $M$ are known and the requirement for a calibration is eliminated. The method then becomes an absolute technique for Pu mass determination.

The variations of measurement and analysis method which are available using the TCA and the interpretation model are summarised below. For each method (except total neutron counting) separate results are obtained for the signal trigger and periodic trigger data capture methods for each of the 16 observation intervals (gate widths).

1. Total neutron counting (1 equation)
   Detection limits

2. Pair Correlation (2 equations)
   2.1 Known $\alpha$ ratio and multiplication $M$
       Unknown $F_S$, $\epsilon$       Method FE
   2.2 Known $\alpha$ ratio and detection
       probability $\epsilon$
       Unknown $F_S$, $M$       Method FM

3. Absolute $^{240}$Pu mass determination
   (3 equations) Triple Correlation
   3.1 Known $\alpha$ ratio
       Unknown $F_S$, $\epsilon$, $M$       Method FEM
   3.2 Known Multiplication (or $M = 1$)
       Unknown $F_S$, $\epsilon$, $S_\alpha$       Method FESA
TABLE 1. Measurement Results of Small Waste Packages.

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<th></th>
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<td>m(Pu)</td>
<td>m(Pu240)</td>
<td>m_c(Pu240)</td>
<td>m_0(Pu240)</td>
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<td>2.83</td>
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<td>49.28</td>
<td>18.94</td>
<td>15.86</td>
<td>33.70</td>
<td>20.28±1.39</td>
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<td></td>
</tr>
<tr>
<td>SUM 1-5</td>
<td></td>
<td>49.28</td>
<td>18.94</td>
<td>15.86</td>
<td>33.70</td>
<td>20.28±1.39</td>
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</tr>
</tbody>
</table>

* = All five packages measured together.

$\alpha = 0.939$ $\uparrow$

$\bar{\alpha} = 1.27\pm0.07$ $\bar{\varepsilon} = 1.17\pm0.50$
3.3 Known Detection probability $\varepsilon$

Unknown $S_{\alpha}$, $F$, $M$ Method SAFM

MEASUREMENTS AND ANALYSIS

Measurements were performed on a series of 220 litre barrels and waste packages originating in a MOX fuel fabrication plant. The barrels contained small waste packages of 1 to 2 kg which had first been measured by the plant operator using pair correlation and a calibration constant.

The measurements were conducted using a 4$\pi$ thermal neutron detector head (waste monitor) with 60 $^3$He detectors with an exponential decay time of 43.4 $\mu$s and an earlier prototype TCA. Both signal and periodic trigger methods were employed and the data was analysed using Methods FE and FESA as the detection probability $\varepsilon$ had to be treated as an unknown as the distribution of Pu in the 220 litre barrel was unknown.

The results of measurements of five waste packages are summarised in Table 1. The operators declared data are listed in columns 3 and 4 based on the small waste package measurements using a high efficiency calibrated in plant detector. Column 5 lists the results based on a calibration of the 220 litre waste monitor using a standard and pair correlation. These results seriously underestimate the declared values.

The results of the TCA measurements using pair correlation (Method FE) are listed in columns 6 and using triple correlation (Method FESA) in columns 7, 8 and 9. The pair correlation analysis assumed that all barrels had the same declared isotopic composition with $\alpha = 0.939$. The resulting significant errors indicate that pair correlation analysis is inappropriate. In contrast, very good agreement with the operators individual package measurements is achieved using (absolute) triple correlation analysis.

The results of the TCA measurements using triple correlation (Method FESA) for 11 barrels and employing the periodic trigger method are tabulated in Table 2 in order of increasing $\alpha$ ratio. Again, very good agreement is achieved with the operators declared values. In the case of high $\alpha$ ratio measurements, the periodic trigger data capture method performed better than the signal trigger method which fails.

Some justification for the superior performance of the periodic trigger data capture procedure where a high proportion of uncorrelated single neutrons are present (high $\alpha$ ratio) can be obtained from an examination of the equations for the total number of measured triplets. The total numbers of measured triplets for the signal trigger method $N_{S(12)}$ and for the random (periodic) trigger method $N_{T(3)}$ for observation interval $\tau_1$ are given by:

$$N_{(12)}(\tau_1) = T_M[R_3 f^2(T, \tau_1) + R_2 f(T, \tau_1) + W_2(\tau_1)] R_1 \tau_1 + 1/2 (R_1 \tau_1)^2 R_1$$

$$N_{T(3)}(\tau_1) = T_M[R_3 W_3(\tau_1) + R_2 W_2(\tau_1) R_1 \tau_1 + 1/6 (R_1 \tau_1)^2 R_1]$$

where $f(T, \tau_1)$ is a function of $T$, $\tau_1$, and the detector decay constant $\lambda$ and where $W_2$ is a function of $\tau_1$ and the detector decay constant.
In both of the above equations the final term on the right hand side corresponds to the uncorrelated triplets count. The influence of this term is significantly less important for the random (periodic) trigger method where the number of uncorrelated triplets is high, for example, due to the presence of a high (α,n) source.

CONCLUSION

Neutron multiple correlation analysis (multiplicity counting) using the TCA provides a versatile and effective range of relative and absolute measurement methods for determining the Pu mass of plutonium bearing waste and bulk plutonium samples. The use of multiple observation intervals and independent data capture methods provides confidence in the measurement results and a degree of measurement validation. The random (periodic) trigger method is particularly suitable for the measurement of plutonium bearing samples where a high proportion of neutrons are uncorrelated, for example material with a high α ratio.

REFERENCES


