DEVELOPMENT STATE OF THE TRIPLE NEUTRON CORRELATION TECHNIQUE BY THE FACTORIAL MOMENT METHOD AT JRC ISPRA

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ABSTRACT

The mathematical models for pair and triple correlation methods based on the factorial moments of the measured frequency distributions are explained. The effects discussed include neutron multiplication and dead time.

1. Introduction

The Joint Research Centre Ispra is working since 23 years in the field of passive neutron assay. 1969 BIRKHOFF and BONDAR started with the development of the variable dead time counter VCD (Ref.1). Later followed a simulation of the ROSSI α - method with the LABEN computer (Ref.2 till 4), which was applied during the early seventies to Pu contaminated reprocessing waste (Ref.5).
A first version of a Time Correlation Analyzer was realized by Mr.BONDAR 1979 using a PLESSEY computer for a fast data acquisition (Ref.6). The data analysis was based on an interpretation model of the probability distibution of signal events in either signal triggered or periodically triggered inspection intervals using a computer simulation (Ref.7 till 9).
Analytical models for the probability distribution and the respective factorial moments were published between 82 and 86 (Ref.10, 11), taking into account neutron multiplication effects by analytical expressions (Ref.12, 13). Tests of the pair correlation interpretation models with the analytical neutron multiplication model were started already during 1984 (Ref.14).
The same formalism for the interpretation of the pair correlation analysis was later obtained by Mr.ENSSLIN (Ref.15) based on a publication of Mr.BOEHNEL (Ref.16).
The first experiments with a fixed wired instrument for triple correlation analysis were performed during early 1987. For this purpose a FORTRAN computer programme is available for the data acquisition elaboration and interpretation.

The formulas used are summarized in Ref.13 and represent the state of knowledge of 1986. The hardware is an analyzer built by the ROGOWSKI Institute of the University of AACHEN (Ref.17) for two different inspection interval types. In Type 1 each detected neutron signal at Interval $[t_1 \quad t_1 + dt_1]$ opens after a delay T at time $t_1 + T$ a set of inspection interval of 15 different durations $\tau_i = i \tau_0 \quad i = 1, 2 \cdots 15$

In type 2 16 inspection intervals of durations are opened periodically without any time gap between these intervals. The correlation analyzer provides such 31 analysis results. The signal resolution is 0.1 µs. This instrument was mainly designed for the assay of radiaoctive waste (Ref.18 till 21). A commercial instrument of the second generation was built at ASEM/UDINE during 1988 using only the Inspection Interval Method of type 1. It has a multiplicity counting capacity of 128 and can be used as well for bulk material. A third generation TCA will be on the market during 1993 and will have all features required for waste and bulk Pu assay.

The present report gives the present state of art of the algorithms developed at the JRC for the interpretation of neutron signal correlation instrumentation.

2. The idealised singlets, correlated pairs and triplets

The pair and triple correlation analysis is based on a point model, in which the correlated multiplets are directly proportional to the spontaneous fission rate F_S . These expressions are :

$$R_1 = \varepsilon F_S M \nu_{S(1)} (1+\alpha) \tag{1}$$

$$R_2 = \varepsilon^2 F_S M^2 \nu_{S(2)} \left[1 + (M-1)(1+\alpha) \frac{\nu_{S(1)} \nu_{I(2)}}{\nu_{S(2)} (\nu_{I(1)} - 1)} \right] \tag{2}$$

$$R_3 = \varepsilon^3 F_S M^3 \nu_{S(3)} \left[1 + 2(M-1) \frac{\nu_{S(2)} \nu_{I(2)}}{\nu_{S(3)} (\nu_{I(1)} - 1)} + (M-1)(1+\alpha) \left[\frac{\nu_{S(1)} \nu_{I(2)}}{\nu_{S(3)} (\nu_{I(1)} - 1)} \right] \left[1 + 2(M-1) \frac{\nu_{I(2)}^2}{\nu_{I(3)} (\nu_{I(1)} - 1)^2} \right] \right] \tag{3}$$

$$M = \frac{1-p}{1-p\,\nu_{I(1)}} \tag{4}$$

$$\alpha = \frac{S_\alpha}{\nu_{S(1)} F_S} \tag{5}$$

$$\nu_{j(\mu)} = \sum_{\mu=\nu} \binom{\nu}{\mu} P_{j\nu} \tag{6}$$

R_μ is the rate of correlated signal multiplets of order μ.

The symbols used are :

S_α = (α,n) neutron emission rate of test item

F_S = spontaneous fission rate of test item

p = probability that a neutron generates an induced fission event

ε = probability for detection of a neutron

$P_{j\nu}$ = probability for the emission of ν fast neutrons per prompt fission caused by a primary neutron generated by reaction j (j = I when induced fission, j = s for spontaneous fission).

$\nu_{j(\mu)}$ = μth factorial moment of the $P_{j\nu}$-distribution

R_1 and R_2 are obtained from the Shift Register, from the Reduced Variance meter and from the Variable Dead Time counter. The Euratom time correlation analyzer gives 16 pairs of R_1 and R_2 for both techniques, the shift register and the reduced variance meter. In addition it can deliver 16 values of R_3, R_4 ... for a generalization of each of the two techniques. However for detection probabilities of $\varepsilon < 0.2$ the R_4- values are heavily influenced by cosmic radiation neutron bursts. The JRC has developed explicit interpretation models which are valid for waste and safeguards applications permitting an analysis of either 2 or 3 unknown parameters of waste or fuel items.
The developed software (Ref.17, 22) permits following applications :

1. Absolute Pu 240 mass equivalent determination knowing the isotopic composition

   Routine : FEM

   Unknown : F_s, ε, M known α

2. Absolute Pu 240 mass equivalent determination for dispersed Pu (MOX waste) $M \simeq 1$

   Routine : FESA

   Unknown : F_s, ε, S_α known M

3. Absolute Pu 240 mass equivalent determination for bulk material and unknown isotopic composition

Routine : SAFM

Unknown : S_α, F_S , M known ε

The routine FEM can be operated as well in the pair correlation mode with α and M as input. The same applies for the routine SAFM. Here the α-ratio and ε or a standard can be used in the input. This is the normal shift register analysis as practised by the inspectors and programmed at the JRC in the PECC software (Ref.23).

3. Measurement of the multiplets

The neutron detector head provided for the measurement of the spontaneous fission neutrons, (α-n)neutrons and induced fission neutrons has a cylindrical cavity for the accomodation of a Pu item. This cavity is surrounded by a polythene moderator with incorporated He3 neutron detectors. 3 to 4 He-3 counters are grouped together and connected to a conventional chain consisting of an amplifier and a discriminator.

The neutron signals of each detector group are collected in a mixer. The mixer output serves as input into the time correlation analyzer which gives the frequency for the occurance of the number of signals obtained in the inspection intervals.

In method 1 each signal of a signal pulse train existing in the time interval (t_1, t_1+dt) opens with a setable delay T at time t_1+T an observation interval and closes at time $t_1 + T + \tau$.

In method 2 the intervals are opened periodically without any rest time from interval to interval. The technical specifications of the third instrument generation are :

Signal frequency synchronization	10 MHz
Basic observation interval	0.2, 0.4, 0.8, 1.6 3.2, 6.4, 12.8, 25.6 µs
Number of observation interval 16	
Predelay settings	0, 0.5, 1. .... 7.5 µs
Input signals	TTL or Optical signals
Highest frequency	16 for waste items 256 for large Pu quantities

The single counts.

The total number of counts can be measured in two different ways. One method uses simply a time gated counter and registers at the mixer output the number of signals $N_{t(1)}$ obtained during a measurement time T_M . This experimental quantity is related to the theoretical by :

$$N_{t(1)} = T_M R_1 \tag{7}$$

In the second method the number of signal events $B_x(\tau_i)$ with x signals in periodically opened observation intervals τ_i are used by forming the first factorial moment. It is

$$N_{b(1)}(\tau_i) = \sum_{x=1}^{\infty} x B_x(\tau_i) \cong T_M R_1 \tag{8}$$

These $B_x(\tau_i)$ events are obtained by $\frac{T_M}{\tau_i}$ opened inspection intervals of duration τ_i without time gap between the intervals. $N_{b(1)}(\tau_i)$must be the same for all 16 inspection interval seizes τ_i, $i = 1, 2, 3 \cdots 16$.

In case that the neutron detectors have an updating neutron dead time δ the relation 7 or 8 is not anymore valid. For one amplifier chain following relation is obtained

$$N_{b(1)}(\delta) = T_M P_0 \sum_{n=1}^{\infty} R_n (1 - e^{-\lambda\delta})^{n-1} \tag{9}$$

with

$$\ln P_0 = \delta \sum_{n=1}^{\infty} (-1)^n R_n w_n(\delta) \tag{10}$$

and

$$w_n(\delta) = 1 + \sum_{k=1}^{n-1} \binom{n-1}{k} (-1)^k \frac{1 - e^{-k\lambda\delta}}{k\lambda\delta} \tag{11}$$

Eq 9 can be applied for many amplifier chains i = 1,2, ...I) working in parallel. This is true if each chain has the same neutron detection probability ε, the same dead time δ and $\lambda\varepsilon << 1$. With these conditions it can be shown that

$$\delta = \frac{\delta_1}{I} = \frac{\delta_2}{I} \cdots \frac{\delta_I}{I} \tag{12}$$

I = number of parallel amplifier chains

Pair counts.

The total number of doublets measured with the time correlation analyzer and the periodically opened inspection intervals reads :

$$N_{b(c)}(\tau_i) = \sum_{x=2}^{\infty} \binom{x}{2} B_x(\tau_i) \tag{13}$$

This measured quantity can be expressed for neutron detection without dead time by (Ref.11,24) :

$$N_{b(c)}(\tau_i) = T_M \left[R_2\, w_2(\tau_i) + \frac{1}{2} R_1 \tau_i R_1 \right] \tag{14}$$

$$w_2(\tau_i) = 1 - \frac{1}{\lambda \tau_i}\left[1 - e^{-\lambda \tau_i}\right] \tag{15}$$

Pair counts $N_{(11)}(\tau_i)$ are obtained in signal triggered inspection intervals from the measured frequency distribution of the signal pulse train.

In this case each signal of a pulse train detected in the time intervals $[t_1, t_1+dt_1]$ triggers i intervals $[t_1+T, t_1+T+\tau_i]$ with $0 \leqq t_1 = T_M \gg T+\tau_i$. It is then :

$$N_{(11)}(\tau_i) = \sum_{x=1}^{\infty} x N_x(\tau_i) \qquad i = 1, 2, 3 \cdots 15 \tag{16}$$

$N_x(\tau_i)$ = number of counts with x signals inside inspection intervals of duration τ_i obtained during a measurement time $T_M \gg \tau_i$. Each signal starts a set of i 1, 2 ... 15 inspection intervals.

The measured pair counts can be set equal to the respective theoretical expression.

$$N_{(11)}(\tau_i) = T_M \left[R_2\, f(T,\tau_i) + R_1\, \tau_i\, R_1 \right] \tag{17}$$

$$f(T,\tau_i) = e^{-\lambda T} \left[1 - e^{-\lambda \tau_i} \right] \tag{18}$$

Eq. 17 is the same expression as obtained for the so called Shift Register. The main difference between the two pair correlation methods exists in the last terms of the Eqs. 14 and 17. The number of uncorrelated signals contributing to the pair counts is greater in the case of the shift register. If the counters have an updating dead time δ then the pair counts $N_{b(2)}(\delta,\tau_i)$ or $N_{(11)}(\delta,\tau_i)$ have a more complex structure. It is according to Ref. 25 :

$$N_{(11)}(\delta,\tau_i) = P_0^2 \Big[a_{21} R_2 + a_{22} R_3 + a_{23} R_1^2 + a_{24} R_1 R_2 + a_{25} R_1 R_3 + a_{26} R_2^2 \Big] \tag{19}$$

The same expression but with different coefficients a_{2j}, $j = 1, 2 \cdots 6$ is valid for $N_{b(2)}(\delta,\tau_i)$.
To the correlated pairs R_2 contribute as well correlated triplets R_3 .
In case that the spontaneous fission and (α, n) reaction rate is very low then all square terms R_1^2 , $R_1 R_2$ .... can be neglected and $N_{11}(\delta,\tau_i)$ is (Ref. 24):

$$N_{(11)}(\delta, \tau_i) = T_M \, e^{-2\delta R_1} \, R_2 \, f(T, \tau_i) \left[1 - 2 \frac{R_3}{R_2} \lambda \delta \left(1 + \frac{f(T, \tau_i)}{2} \right) \right] \qquad (20)$$

$$\text{for } \lambda \delta \ll 1$$

This shows that even for small spontaneous fission rates there exists a dead time correction which can only be neglected if following condition is fullfilled :

$$3 \lambda \delta \varepsilon \ll 1 \qquad (21)$$

Triple counts

Periodically opened inspection intervals lead to the following number of measured triplets

$$N_{b(3)}(\tau_i) = \sum_{x=3}^{\infty} \binom{x}{3} B_x(\tau_i) \qquad (22)$$

The theoretical counterpart to these measured number of triplets is given by (Ref.11,24):

$$N_{b(3)}(\tau_i) = T_M \left[R_3 \, w_3(\tau_i) + R_2 \, w_2(\tau_i) \, R_1 \tau_i + \frac{1}{6} (R_1 \tau_i)^2 R_1 \right] \qquad (23)$$

$$w_3(\tau_i) = 1 - \frac{1}{2 \lambda \tau_i} \left[3 - 4 e^{-\lambda \tau_i} + e^{-2 \lambda \tau_i} \right] \qquad (24)$$

The corresponding expressions for the signal triggered observation intervals are (Ref..11,24):

$$N_{(12)}(\tau_i) = \sum_{x=2}^{\infty} \binom{x}{2} N_x(\tau_i) \qquad (25)$$

and

$$N_{(12)}(\tau_i) = T_M \left[R_3 f^2(T,\tau_i) + R_2 \left[f(T,\tau_i) + W_2(\tau_i) \right] R_1 \tau_i + \frac{1}{2}(R_1 \tau_i)^2 R_1 \right] \tag{26}$$

Comparing Eq 26 with 23 it is clear that both methods have their merits. Correlated triplets R_3 obtained with the signal trigger method of Eq.26 suffer more under uncorrelated triplets caused by the combination of singlets with correlated doublets and by singlets. Such an occasion exists if waste with high α-ratios has to be assayed with the triple correlation method (Ref.21). In order to demonstrate which method leads to better results it is necessary to elaborate first a detailed error model. In case that the signal pulse train is measured with an updating dead time δ then $N_{b(3)}(\delta,\tau_i)$ and $N_{(12)}(\delta,\tau_i)$ can be approximated by following expression (Ref.25) :

$$\begin{aligned} N_{(12)}(\delta,\tau_i) = P_0^3 \Big[& a_{31} R_3 + a_{3,12} R_4 \\ & + a_{32} R_1 R_2 + a_{33} R_2^2 + a_{34} R_1 R_3 + a_{35} R_2 R_3 \\ & + a_{36} R_1^3 + a_{37} R_1^2 R_2 + a_{38} R_1^2 R_3 + a_{39} R_1 R_2^2 + a_{3,10} R_1 R_2 R_3 + a_{3,11} R_2^3 \Big] \end{aligned} \tag{27}$$

The coefficients a_{31} for $N_{(12)}(\delta,\tau_i)$ are tabulated in Ref.24 and those for $N_{b(3)}(\delta,\tau_i)$ in an internal JRC Report. The none linear equations for $N_{b(1)}(\delta)$, $N_{b(2)}(\delta,\tau_i)$, $N_{b(3)}(\delta,\tau_i)$ and $N_{(1j)}(\delta,\tau_i)$ $j=1,2$ can be solved by iterative methods to obtain the singlets R_1, the correlated doublets R_2 and the correlated triplets R_3.

4. Developments

Based on the extensive development of the neutron correlation technique at JRC Ispra, a new third generation neutron Time Correlation Analyser is being developed by ANTECH under licence from the Institute for Safety Technology of the JRC. The instrument is based on a flexible electronic design with configurations for both waste and bulk material measurement.

The specification provides for both periodic and signal triggered multiple observation intervals and the recording of neutron multiplicities ranging from 16 for waste to 256 for bulk material. Extensive diagnostic and test facilities have been incorporated into the design.

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