ANS 1993 Annual Meeting & ANS Topical Meeting on

National Critical Technologies - Advanced Technology Applications

June 20 - 24, 1993

San Diego, California

TRIPLE NEUTRON CORRELATION FOR MOX WASTE ASSAY

W. Hage, B. Pedersen, L. Bondár
Commission of the European Communities, Joint Research Centre Ispra,
Safety Technology Institute, 21020 Ispra (Va), Italy.

M. Swinhoe
Commission of the European Communities, Safeguards Directorate, Luxembourg.

F.W. Ledebrink
Siemens AG, Hanau, Germany.

Introduction.

Triple neutron signal correlation techniques were applied to measure the waste in 220 litre barrels. Small waste packages of 1 to 2 kg weight originating from a MOX fuel fabrication plant were first measured in a dual range detector head by the plant operator. The correlated pair count rate of each individual package was used together with a calibration constant to determine the effective ^{240}Pu mass equivalent. A set of such packages was then loaded into a 220 litre barrel keeping the α activity well below a prescribed tolerance limit. The task was to verify the ^{240}Pu equivalent mass content in each barrel for safeguards purposes.

Method.

The 220 litre waste barrels were measured inside a 4π thermal neutron detector head with 60 ^{3}He neutron detectors having an exponential response function over more than two decades with $1/\lambda$ = 43.4 μs. The signal pulse train generated by the spontaneous fission neutrons of the Pu isotopes with even mass number and the neutrons from (α,n) reactions were analyzed in a frequency analyzer in two distinct ways. In the first method the discrete frequency distribution p_x, x = 0,1,2,3,... in periodically opened inspection intervals $[t_1,t_1+\tau]$, $[t_1+\tau,t_1+2\tau]$, ..., $[t_1+(k-1)\tau,t_1+k\tau]$ were measured. The measurement time T_M is equal to $k\tau$. In the second method the bivariate frequency distribution p_{x1x2} was registered for $x_1 = 1$ and $x_2 = 0,1,2,...,\infty$ i.e. each signal observed in the interval $[t_1,t_1+dt_1]$ opens an inspection interval $[t_1+T,t_1+T+\tau]$ with a delay T during the total measurement time $0 \le t_1 \le T_M$.

A theory was developed in which the normalized factorial moments of the probabilities p_x and p_{x1x2} in contrast to earlier work[1] could be expressed in closed form as function of physical, nuclear and instrumental parameters by means of the exponential Bell polynomials B_{nk}

$$m_n = \sum_{x=n}^{\infty}\binom{x}{n}p_x = \frac{1}{n!}\sum_{k=1}^{n}B_{nk}(D_1,D_2,\ldots,D_n) \qquad (1)$$

and

$$m_{1n} = \sum_{x_1=1}\sum_{x_2=n}^{\infty}\binom{x_1}{1}\binom{x_2}{n}p_{x_1x_2} = \frac{1}{1!\cdot n!}\left[D_{1n} + \sum_{v=1}^{n}\binom{n}{v}D_{1(n-v)}\sum_{k=1}^{v}B_{vk}(D_{01},D_{02},\ldots,D_{0v})\right] \qquad (2)$$

It is:

$$D_n = D_{0n} = n!\,d_n\,\tau\,w_n(\tau) \qquad (3)$$

$$w_n(\tau) = 1 + \sum_{k=1}^{n-1}\binom{n-1}{k}(-1)^k\frac{1-e^{-\lambda k\tau}}{\lambda k\tau} \qquad (4)$$

$$D_{1n} = n!\,d_{n+1}\,T_M\,f^n(\tau) \qquad (5)$$

$$f(\tau) = e^{-\lambda T}\left(1-e^{-\lambda\tau}\right) \qquad (6)$$

$$d_n = \varepsilon^n \left[F_S \nu_{s(n)} + S_\alpha (1 - \delta_{1n}) \right] \quad (7)$$

F_S = spontaneous fission neutron emission rate,
S_α = (α,n) neutron emission rate,
ε = detection probability,
T_M = measurement time,
T = pre-delay,
τ = observation interval,
λ = decay constant of thermal neutron detection assembly,
P_{sv} = probability distribution for emission of ν spontaneous fission neutrons.

$$\nu_{s(n)} = \sum_{\nu=n}^{\infty} \binom{\nu}{n} P_{s\nu} \quad (8)$$

From the frequency distributions the factorial moments m_1, m_2 and m_3 in method 1 and m_{10}, m_{11} and m_{12} in method 2 were used to determine from the eqs. 1 to 8 the unknowns F_S, S_α and ε. The detection probability ε had to be considered as an unknown due to the undefined distribution of Pu in the 220 litre barrel. The decay constant λ was determined in a separate experiment. The developed software includes as well the branching process of neutron multiplet multiplication.

Results.
A set of small samples with waste was measured first in the dual range detector head of the plant operator then in the JRC detector head. These packages were made up of MOX powder residues sticking to the walls of Al containers wrapped in plastic.
To distinguish in table 1 one package from the other they were named for example KT8410 if there were 8410 mg Pu declared[2]. The corresponding ^{240}Pu mass equivalents are listed in column 4. Column 5 gives the $m_c(^{240}Pu)$ values calibrating the 220 litre waste barrel monitor with a standard and the correlated neutron signal pairs. This procedure underestimated the declared masses between 14 % and 20 % with one exception. Column 6 gives the results of pair correlation $m_p(^{240}Pu)$ under the assumption that all waste packages have the same isotopic composition with an α of 0.939. This value is far too low. For this reason pair correlation can not be used. Triple correlation with (18.77±1.6)g ^{240}Pu equivalent agrees very well with the sum of the assay values measured with the plant operators instrument, where each of the 5 packages were measured individually (18.94 g).
Table 2 summarizes the obtained ^{240}Pu mass equivalent values of the 11 barrels measured, ordered with increasing α-ratio. Each barrel was measured three times 15 minutes. This measurement time was sufficient for α-ratios below 3. Barrels with larger ^{241}Am contamination e.g. barrel no. 11 require much longer measurement times to obtain a similar precision.

Conclusions.
The plant operator data obtained by small waste package measurements based on a calibration model could be confirmed with the triple correlation being an absolute method. In addition the α contamination related to the α-value of the waste can be determined. Individual barrels with an α value below 2 are measured with an accuracy below 10 %. For higher α-values the assay results can be improved by the implementation of the dead time corrections[3] and using an optimum moderator to detector volume ratio in the detector head.

References.

1. W. HAGE and D.M. CIFARELLI, "Correlation Analysis with Neutron Count Distributions in Randomly or Signal Triggered Time Intervals for Assay of Special Fissile Materials", *Nucl. Sci. Eng.*, **89**, 159-176 (1985).

2. L. BONDÁR et. al., "Pu Waste Measurements in a MOX Fuel Fabrication Plant by Neutron Correlation Methods", EUR Report (to be published 1993).

3. W. HAGE and D.M. CIFARELLI, " Correlation Analysis with Neutron Count Distributions for a Paralyzing Dead-Time Counter for the Assay of Spontaneous Fissioning Material", *Nucl. Sci. Eng.*, **112**, 136-158 (1992).