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§1. Introductory Remarks

This paper addresses problems of calibration and estimation of a parameter, p_0 , in high level neutron coincidence counting (HLNC) measurement. The presentation begins with a deductive approach, based on the generally accepted physical equations which are satisfied by the total and coincidence count rates^{1,2}. A new approach for the estimation of p_0 via measurement and a new calibration procedure consistent with the physical theory of coincidence counting measurement^{1,2} are presented. The approach is compared with the existing conventional approach in which possible inconsistency with the theoretical model is shown.

In addition, the original model for neutron coincidence counting in a multiplying medium was extended to simultaneously account for the possible presence of poison, as well as for the neutron detection.³ The equations suitable for the data analysis of measurement of waste and their criteria are given.

§2. Theoretical Considerations

The basic equations underlying the use of HLNC for the determination of the plutonium content of samples are^{1,2}

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- 1 K. Böhnel, "The Effect of Multiplication on the Quantitative Determination of Spontaneously Fissioning Isotopes by Neutron Correlation Analysis", Nuclear Science and Engineering, 90, (1985), pp.75-82.
 - 2 W. Hage and D. M. Cifarelli, "On the Factorial Moments of the Neutron Multiplicity Distribution of Fission Cascades", Nuclear Instruments and Methods in Physics Research, Section A, A 236, (1985)pp.165-177.
 - 3 M-S. Lu and T. Teichmann, "A Generalized Model for Coincidence Counting", to appear in Nuclear Instruments and Methods in Physics Research, Section A.

$$T = \epsilon M v_{1s} (1 + \alpha) C m \quad (1)$$

$$R = \frac{1}{2} \epsilon^2 M^2 f \left[v_{2s} + \frac{(M-1)v_{1s}}{v_{1i} - 1} v_{2i} (1 + \alpha) \right] C m \quad (2)$$

- Here
- T = totals count rate;
 - R = reals coincidence count rate;
 - ϵ = detection efficiency;
 - v_{1s} = first moment, $\langle n_s \rangle$, of the spontaneous fission neutron emission distribution function, about 2.14;
 - v_{2s} = second factorial moment of the spontaneous fission distribution function (i.e. the mean value $\langle n_s(n_s - 1) \rangle$ of the spontaneous fission neutron emission distribution function, about 3.735;
 - v_{1i} = first moment of the induced fission distribution function, about 2.876;
 - v_{2i} = second factorial moment of the induced fission distribution function, about 6.748;
 - α = ratio of the neutrons from (α, n) reactions to those from spontaneous fission;
 - m = effective mass of Pu^{240} ;
 - C = rate of spontaneous fissions of Pu^{240} , about 475 /g/s;
 - f = fraction of neutrons counted within the coincidence gate,

$$f = e^{-d/\tau} (1 - e^{-g/\tau}) \quad (3)$$

where τ is the detector die-away time, d the pre-delay and g the coincidence gate width;

and M = leakage multiplication factor, given by

$$M = \frac{1 - p_f}{1 - p_f v_{1i}} \quad (4)$$

where p_f is the probability of inducing fission.

The underlying assumptions in the model are that each neutron in the sample has the same probability to induce a fission and that neutrons with the same pedigree can register a coincidence pair during measurement. The expressions were initially derived independently by Böhnel¹ and Hage and Cifarelli² considering explicitly only the effect of induced fissions in the sample.

Recently a more physically transparent model was developed. The model accounts simultaneously and explicitly for the effects of induced fissions, poison, as well as instrumental detection. In particular, it was shown that the detection efficiency, ϵ , in equations (1) and (2) above should be expressed as the product of an intrinsic detector efficiency, ϵ_0 , the probability that a neutron impinging on the detector system will register a count in the detector, and the probability that a neutron generated in the sample will reach the detector. In terms of the probabilities of each neutron to induce a fission, p_f and the capture probability, p_c , i.e. the probability of being absorbed (for example, by a poison) without inducing a fission, the detection system efficiency can then be expressed as

$$\epsilon = \epsilon_0 \frac{1 - p_f - p_c}{1 - p_f} \quad (5)$$

§3. A New Calibration Procedure

From equations (1) and (2), calibration is viewed as a procedure to determine the detector efficiency, ϵ , and the fraction of neutrons counted within the coincidence gate-width, f . The properties of the material used in calibrations are assumed known. The calibrated parameters are then used to assay the plutonium content of unknown samples.

Firstly, the detector die-away time for each sample is determined by measuring the coincidence count rates by varying the gate-widths. For example, when the coincidence count rates corresponding to gate-widths of g_1 and g_2 , where $g_2 = 2g_1$, are R_1 and R_2 , then

$$\tau = -g_1 / \ln(R_2/R_1 - 1). \quad (6)$$

With this value of τ and the known pre-delay d and gate-width g , the fraction of neutrons counted within the coincidence gate is determined for each sample. (Approximate values for the HLNCII are: $\tau=43\mu s$, $d=4.5\mu s$, $g=64\mu s$ and thus $f=0.697$.)

Secondly, the detection efficiency for each sample in the detector during calibration is determined. This is achieved by eliminating ϵM from equation (1) and then solving equation (2) for M , which yields

$$M = \frac{2Rv_{1s}^2 (1 + \alpha)^2 C_m - [1 - k(1 + \alpha)] T^2 v_{2s}}{k(1 + \alpha) T^2 v_{2s}} \quad (7)$$

Here k is a physical constant of the system

$$k = \frac{v_{1s} v_{2i}}{(v_{1i} - 1) v_{2s}} \quad (8)$$

which occurs both in equation (2) above and in the quadratic equation determining M which is used in a variant of the procedure (see equation (12) below).

The detection efficiency is then obtained from

$$\epsilon = \frac{T}{v_{1s} (1 + \alpha) C_m M} \quad (9)$$

With ϵ and f for each sample so determined, a statistical computational procedure is then used to obtain an optimum set of ϵ^* and f^* for subsequent applications. If the material to be assayed is similar to the calibration standards, these parameters should not change much between calibration and applications. Various criteria selected by the user, for example, minimum relative variances, minimum absolute total differences (or other criteria) may be used in this statistical process. The statistical variation of ϵ and f during the calibration process provide an estimate of the accuracy and precision to be expected during applications.

The application of the ϵ^* and f^* obtained to determine the plutonium mass is trivial. Similar to conventional HLNC data analysis, the value of α for each sample is assumed known. Equations (1) and (2) are then used to solve for the two unknowns, M and m .

§4. The Conventional Procedure

The conventional algorithm for HLNC data analysis is obtained by first multiplying equation (2) by $(1+\alpha)$ and then dividing it by equation (1) to get a quadratic equation in M :

$$\frac{R(1+\alpha)}{T} = \frac{1}{2} \epsilon f M \frac{v_{2s} + \frac{(M-1)v_{1s}}{v_{1i} - 1} v_{2i} (1+\alpha)}{v_{1s}} \quad (10)$$

Noticing that the resulting expression still contains the unknown factors ϵ and f on the right-hand side, Ensslin⁴ suggests bypassing this problem by introducing the quantity p_0

$$p_0 = \left[\frac{R(1+\alpha)}{T} \right]_0 \quad (11)$$

where the subscript "0" denotes a non-multiplying sample, for which $M=1$. The Ensslin equation for M is then obtained by dividing equation (10) by p_0 to arrive at:

$$k(1+\alpha)M^2 + [1 - k(1+\alpha)]M - r = 0 \quad (12)$$

where k is a physical constant of the system, defined above (see equation (8)), and

⁴ N. Ensslin, "A Simple Self-Multiplication Correction for In-Plant Use", Proceedings of the 7th Annual ESARDA Symposium on Safeguards and Nuclear Materials Management, (Liege, Belgium, 21-23 May, 1985), ESARDA 19, (European Safeguards Research and Development Association, Brussels, Belgium, 1985) pp.233-238.

$$r = \frac{R (1 + \alpha)}{p_0 T} \quad (13)$$

m, the effective plutonium-240 mass, is then defined by

$$R_c = \frac{R}{Mr} = \frac{p_0 T}{M (1 + \alpha)} = p_0 \epsilon v_{1s} C_m \equiv a_0 m. \quad (14)$$

This last equation defines the conventional calibration constant, a_0 , for the HLNC. Notice that from equations (1) and (2) the quantity p_0

$$p_0 = \left[\frac{R (1 + \alpha)}{T} \right]_0 = \frac{1}{2} \epsilon f \frac{v_{2s}}{v_{1s}} = p_0 (\epsilon, f) = p_0 (\epsilon f) \quad (15)$$

is a function of ϵ and f . In order to minimize the changes to the existing conventional algorithm for the calculation of M when the new procedure is used, the quantity p_0 can be calculated for the measurement system at the calibrated condition, ϵ^* and f^* according to this equation, though its introduction and application are not essential.

If the value p_0 at the measurement condition (i.e., ϵ and f) can be determined, the material properties M and m which are independent of the detector parameters ϵ and f can also be determined. The new calibration procedure described in the previous section aims to achieve this by using calibration standards with material properties similar to the materials to be assayed, and thus assumes that the ϵ and f so determined will not vary greatly in the further applications.

In the standard approach⁴, p_0 is presumed to be obtained by measurement of R and T for small masses (10-20g), or *via* measurements of a series of samples of the material with decreasing masses and eventual extrapolation of the ratio R/T to zero mass. However, references regarding such experimental determination of p_0 have not been published and the practice is not usually exercised. For example, the zero mass measurement is especially difficult for the Universal Fast Breeder Reactor Subassembly Counter (UFBC) or other fuel assembly coincidence counters. Such measurements may prove particularly difficult due to the much lower signal-to-noise ratio for small samples, as well as the much greater statistical fluctuation of the ϵ and f

values for such samples relative to the larger masses encountered in practice. The ϵ and f obtained for small masses may thus deviate greatly from those for larger mass systems of interest.

In fact, a Monte-Carlo simulation on a mainframe computer is usually performed to obtain the value of p . For HLNCII, the value 0.103 was obtained, and this is frequently used in measurements with HLNCII. However, Menlove⁵ has noted that in order to match M between experiment and Monte-Carlo simulation, a value of 0.108 works better. Hybrid methods, combining Monte Carlo techniques with other aspects of the conventional models have also been applied, yielding values of p_0 which vary, sometimes substantially, from 0.103.⁵ It also turns out that the values so obtained may deviate greatly from that obtained in real applications, leading to undesirable inconsistencies.

In the usual measurement procedure for HLNCII, a constant value of $p_0 = 0.103$ is used, regardless of the calibration parameter a_0 actually measured. From equations (14) and (15) one sees that

$$a_0 = \frac{1}{2} \epsilon^2 f C v_{2s} = \frac{2 C v_{1s}^2}{f v_{2s}} p_0^2 \quad (16)$$

Thus, deviation of a_0 from its nominal value shows that ϵ or f (or equivalently p_0) also so deviate, and this should be noted in the further application of the results.

§5. Discussion

In the current phenomenological approach a_0 is determined using the fixed (assumed) value of p_0 by calculating M from equation (12), using the definition (13) of

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- ⁵ H. O. Menlove, "Calibration and Error Reduction in Neutron Coincidence Counting", Nuc. Mater. Manage., XVI, #1 (1987) pp.20-28.
- ⁵ J. E. Stewart, "A Hybrid Monte Carlo/Analytical Model of Neutron Coincidence Counting", Trans. Am. Nuc. Soc., 53, (1986), pp.149-151, and
J. E. Stewart, *et al.*, "Calibration Parameters from Monte Carlo Simulations of Neutron Coincidence Assay of MOX Fuel Elements: A Substitute for Standards?", Proceedings of the 11th Annual ESARDA Symposium on Safeguards and Nuclear Materials Management, (Luxembourg, 30 May - 1 June, 1989), ESARDA 22, (European Safeguards Research and Development Association, Brussels, Belgium, 1989) pp.135 - 141.

r, and finally obtaining a_0 using equation (14). This procedure yields a satisfactory result for the actual case used, and for other measurements for which the efficiency ϵ , the gate-width factor f , and hence the value of p_0 do not change greatly from their nominal values.

Even if the (real) values of p_0 do vary somewhat, this procedure will probably prove adequate, provided that the leakage multiplication factor, M , remains near 1, the point being that

$$\begin{aligned} \frac{dM}{M} &= - \left[\frac{r}{r + M^2 k (1 + \alpha)} \right] \frac{dp_0}{p_0} \\ &\approx -0.3 \frac{dp_0}{p_0} \end{aligned} \quad (17)$$

using results derived elsewhere⁶, i.e. M is not sensitive to changes on p_0 . In addition, equation (14) shows that the calibration parameter is effectively

$$\frac{p_0}{a_0} \quad (18)$$

so that errors in p_0 may be compensated for by countervailing errors in a_0 . (However this possibility needs to be verified by the method described above.) On the other hand, if the samples are such that because of their configuration, their constitution and the presence of impurities, the value of M is substantially changed, then it becomes necessary to use the relation (16) and the equations from which it is derived to obtain a consistent pair of values of p_0 and a_0 to calculate the correct values of the Pu^{240} effective mass m .

More careful study of the physical and mathematical relations cited above shows that there are features which may further mitigate the inconsistencies that may arise in the standard approach. Equation (15) shows that p_0 is a function of the product

⁶ "Analysis of Error Propagation in Gamma Spectroscopy (HRGS) and High Level Neutron Coincidence (HLNC) Counting as Applied to the Non-Destructive Analysis (NDA) of Plutonium-Bearing Materials" Ming-Shih Lu and Theodor Teichmann, (Preliminary Draft) February 1991.

ϵf , and not of ϵ and f separately. If the efficiency, ϵ , is a decreasing function of the neutron energy, while the gate-width factor, f , increases as the neutrons slow down⁷, then their product, and hence p_0 , may not vary greatly from case to case. If confirmed, the standard approach will be applicable in a wide variety of circumstances to a reasonable degree of approximation.

§6. Examples

In order to examine the application of the new procedure described in §3 above, the data in Table VI of LA-10815-MS⁸ for Sellafield PuO₂ samples obtained with the Euratom/LANL HLNCII counter were used. Since the detector die-away time, and hence the gate-width fraction f for each sample are not known, a constant value $f = 0.678$ was assumed. (To be rigorous, this assumption should be checked by measurement of the die-away time for each sample.) Samples #12, 17, and 19 were excluded as described in the document.

For the remaining 16 samples, a calibration constant $a_0 = 18.17$ was obtained when $p_0 = 0.103$ was used in the conventional calibration procedure. It is to be noted that near the end of the same report, a new value of $a_0 = 18.02$ was cited based on additional Cf source measurements to account for sample-container effects and source normalization. This indicates a reduction in efficiency, as can be seen from equation (15). Using the new procedure values of $p_0 = 0.1026$ and $a_0 = 18.08$ were obtained, and the lower efficiency is directly indicated in this approach. The new procedure thus correctly identified the change in the calibration constants from the original data, without the need for the extraneous confirmatory measurements using the Cf source. The new procedure also identified the three cited outliers easily because their efficiencies varied more than four standard deviations from the average of the remaining 16 samples.

⁷ Both of these effects are plausible contributors to the underlying instrument behavior.

⁸ H. O. Menlove *et al.* "Field Test and Calibration of Neutron Coincidence Counters for High-Mass Plutonium Samples", LA-10815-MS, UC-15, February 1987 (Los Alamos National Laboratory, Los Alamos, New Mexico, U. S. A.).

§7. Application to Waste

Interpretation of coincidence measurements for waste are more complicated due to the possible inhomogeneity in the distribution of sources and the presence of neutron absorbers or moderators in the waste. A self-interrogation⁹ method based on empirical correlations between the fissile mass and the ratio of an inferred induced coincidence counting rate over the totals counting rate has been used for the data analysis. Fast coincident counting including triples has also been suggested.¹⁰ Despite the simplicity of the model for coincidence counting, it can be used to assess validity of various approximations and to gain further insights to the problem.

Together with equation (5) for the detection efficiency, equations (1) and (2) can also be used to analyze measurement data for waste. However, the introduction of the additional unknown capture probability due to poison in the waste renders the applications of coincidence measurements more difficult. Nevertheless, when the samples are similar in character to the calibration standards (in that the detection efficiencies and die-away time do not vary significantly between samples and standards measurements), the calibration procedure presented earlier can be used to determine the detection efficiency. In particular, the values of α are assumed known for the standards, the leakage multiplication factors M_s are determined first from equation (7), and then the probabilities to induce fission, p_f , are calculated from equation (4). The detection efficiency ϵ is estimated from equation (9).

In application, as in the case of nominal materials, an estimate of the (α, n) source rates is needed. For waste, neutron sources are mostly likely to be dominated by the americium (α, n) reactions. Fortunately, as can be seen from equations (1) and (2), the leakage multiplication factor M as determined from the measured ratio of R/T is only weakly dependent upon α for large α for which the α dependent terms dominate T and R in these equations. In other words, when $\alpha \gg 1$ and

⁹ P. A. Russo, H. O. Menlove, *et. al.*, "Evaluation of the Neutron Self-Interrogation Approach for Assay of Plutonium in High α -n Materials", Proceedings of the Third International Conference on Facility Operations - Safeguards Interface, November 27 -December 4, 1987, San Diego, California (American Nuclear Society, Inc., La Grange Park, Illinois), p.176.

¹⁰ J. B. Wachter, E. L. Adams and N. Ensslin, "Prototype Fast Neutron Counter for the Assay of Impure Plutonium", *Ibid.* p.194.

$$\alpha \gg \frac{v_{2s}(v_{1i} - 1)}{(M - 1)v_{1s}v_{2i}} - 1$$

the leakage multiplication factor can be determined via

$$\frac{R}{T} = \frac{\epsilon f M (M - 1) v_{2i}}{2(v_{1i} - 1)} \quad (19)$$

The errors in α will not affect M significantly. Substituting this M into equation (1), the error in the measured Pu mass depends linearly on the error in α .

On the other hand, when the fissile content in the waste is small so that the effect of multiplication on the real coincidence count rate R is negligible, or when

$$1 + \alpha \ll \frac{v_{2s}(v_{1i} - 1)}{(M - 1)v_{1s}v_{2i}}$$

the real counting rate alone (equation (2) with M approximately 1) can be used to determine the Pu content. This assumes that the detection efficiency does not deviate significantly from calibration conditions. Using the estimated Pu mass, the value of α can then be obtained from the totals count rate, equation (1), and checked for consistency.

§8. Conclusions

A method has been presented which determines and calibrates the HLNC instrumental parameters, such as the detection efficiency, the gate-width coincidence factor, and the measurement constants ρ_0 and a_0 , for coincidence counters in which the multiplication must be taken into account, based on the measurement of samples with well characterized samples, using the underlying physical equations. This method does not need zero mass measurement, nor the extrapolation of data from small mass measurement, nor does it require a Monte Carlo simulation to match the measurement. When the calibration samples are chosen judiciously, the estimated parameters are more pertinent to the associated inspection situations.

The method can be implemented easily, and should be tested by more actual measurements. In any case, it removes a possible theoretical inconsistency in the conventional procedure.

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