Doubles Counting of Highly Multiplying Items in Moderating, Reflective Surroundings - 11548

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ABSTRACT

When neutrons are counted from a spontaneously fissile self-multiplying item while it is in a reflecting environment, the temporal behavior of the correlated signal following neutron birth is complex. At early times the signal is dominated by prompt fission events coming from spontaneous fission bursts and also from prompt fast-neutron induced fission events. At later times, thermal neutrons 'returning' from the surroundings induce fission and give rise to an additional chain of correlated events. The prompt and returning components probe the fissile and fertile constituents of the item in different ways and it is potentially beneficial to exploit this fact since individual isotopes exhibit different temporal behavior. In this work we look at how the two components can be represented using a linear combination of two simple functions. Fitting of the composite function to the capture time distribution represents one way of quantifying the proportion of each contribution.

Another approach however is to use a dual shift register analysis where after each triggering event two coincidence gates are opened, one close to the trigger that responds preferentially to the prompt dynamics and one later in time which is more sensitive to the returning neutron induced events. To decide on the best gate positions and gate widths and also to estimate the counting precision we can use the analytical fit to work out the necessary gate utilization factors which are required in both these calculations. Here we develop the approach. Illustrative examples are given using spent Low Enriched Uranium (LEU) Pressurized light Water Reactor (LWR) fuel assemblies submersed in borated water and counted in two concentric rings of ^{3}He gas-filled proportional counters. In this case the prompt component (for the range of burnups and cooling time studied) is dominated by ^{244}Cm spontaneous fission and fast-neutron induced fission in ^{238}U for example, while the returning low energy neutrons induce fission mainly in the fissile nuclides such as ^{239}Pu, ^{241}Pu and ^{235}U. One requirement is to calculate the Random Triggered Interrogation Gate Utilization Factor needed to make *á priori* precision estimates but not available from the output of the Monte Carlo simulation code, Monte Carlo N-Particle eXtended (MCNPX).

INTRODUCTION

In nuclear safeguards and waste management, Passive Neutron Multiplicity Counting (PNMC) using Multiplicity Shift Register (MSR)pulse train analysis is widely used to nondestructively quantify Pu. The usual approach is to construct a neutron detector consisting of ^{3}He filled cylindrical proportional counters embedded in a high density polyethylene moderator which surrounds the item. Fast neutrons from the item enter the moderator and are slowed down, on timescales of the order of 1-2 μs, creating a thermal population which then persists typically for several tens microseconds and is sampled by the ^{3}He detectors. Because the slowing down time is short in duration compared to the characteristic thermal lifetime and because the detectors respond predominantly to thermal neutrons (epi-thermal neutrons also contribute to the detected signal when high fill pressures are used but to a relatively small degree) the initial transient can usually be ignored so that the experimentally observed event triggered capture time distribution can be represented by a simple sum of one to three exponentials. This allows simple expressions for the various Signal Triggered Interrogation (STI) and Random Triggered Interrogation (RTI) Gate Utilization Factors (GUFs) to be obtained. These factors represent the proportion of time correlated events present on the pulse train that is actually detected in the coincidence gate [1].

Applying passive neutron multiplicity methods to highly multiplying items adds complexity because neutrons which have emerged from the item may subsequently return to induce fission resulting in signal build-up to the capture time distribution. Fast neutrons that slow down in the item and subsequently induce fission also adds in s similar way. For moderating items the interaction of thermal neutrons within it also contribute to the delayed (non-prompt) signal. An example of such a highly multiplying item (multiplication factors in the range 1.5 to 3) within a closely coupled reflective configuration is the measurement of a spent fuel assembly under water in a neutron collar. This is in contrast to the usual ‘point-model’ assumption of instantaneously multiplication giving rise to an emerging neutron burst that can be considered a ‘super fission’ event [2].

In the case of routine spent commercial pressurized water reactor nuclear fuel assemblies, after only a short cooling time (e.g. 18 months), ^{244}Cm spontaneous fission dominates the initiation process for the creation of correlated neutrons. The signal is considerably increased by multiplication. To manage random pile-up (so called ‘Accidentals’), detectors with short temporal characteristics, relative to the dynamics of the neutron fission chain, are needed so that a gate width as short as possible can be used while retaining a reasonably high GUF. A short dieaway can be achieved by using a high concentration of high pressure ^{3}He proportional counters embedded in a compact moderating assembly clad in, and possibly also containing a thermal neutron poison such as Cd in the form of fins between the proportional counters. Such a configuration effectively tracks the time evolution of the fission rather than defining the capture time distribution.

In this work we show how for a detector deliberately designed to take advantage of this time behavior the capture time distribution with a spent fuel assembly present can be represented by the sum of a prompt and a returning (or reflected) neutron component. GUFs needed for *á priori* precision estimates can then be extracted. As a reminder, the standard deviation σ_D on Doubles counting rates may be approximated from predicted Singles and Doubles rates, S and D respectively, as follows [3]:

$$\sigma_D = \sqrt{1 + 8.g_d.\frac{D/f_d}{S}}.\sqrt{\frac{D + 2.S^2.T_g}{t}}$$

where T_g is the width of the coincidence gate and t is the assay period. f_d and g_d are the STI and RTI GUFs respectively. Equations of this kind are important in optimization studies [4].

CAPTURE TIME DISTRIBUTION MODEL

We use a simple functional form to describe the capture time distribution $f(t).dt$. The capture time distribution is defined as the fraction (probability) of neutron events in the time interval dt about t . It is made up of two contributions. The first, what we shall refer to as the prompt contribution, with normalized shape $P(t).dt$, which one can imagine represents the behavior of fast neutrons born and interacting in the fuel assembly on a time scale which is short compared to the lifetime of neutrons in the detector so that the capture time distribution can be modeled as an instantaneous rise followed by the sum of decaying exponentials [1]. The second contribution with normalized shape $R(t).dt$ we refer to as the reflected or returning contribution and can be thought of as the signal that arises from fissions induced by neutrons that have emerged from the fuel assembly to return at a later time. The shape of this distribution first rises to a peak before dropping off [5]. These designations are somewhat simplistic in meaning but serve for the present purpose to distinguish fast and thermal neutron contributions. The important thing is that together the combined form allows us to fit real or simulated capture time profiles empirically. Algebraically we have:

$$f(t).dt = S.[\alpha.P(t).dt + \beta.R(t).dt]$$

with

$$\beta = 1 - \alpha$$

where α is the fraction of events in the prompt component, β is the corresponding fraction in the returning contribution, and the scale factor S accommodates actual data which may not be normalized over all time $t \geq 0$. For the prompt temporal component we adopt a two time constant exponential decay model as follows [1]:

$$P(t).dt = w_A.e^{-\lambda_A.t}.\lambda_A.dt + w_B.e^{-\lambda_B.t}.\lambda_B.dt$$

with

$$w_A = 1 - w_B$$

where the $\frac{1}{e}$ time constant for component A are given by $\tau_A = {}^{1}/_{\lambda_A}$ and the fraction of events in $P(t)$ that are in component A is w_A. An obvious extension of notation explains the meaning of the B terms.

The returning distribution has the following parameterization:

$$R(t).dt = w_1.\frac{\lambda_0}{(\lambda_0 - \lambda_1)}.[e^{-\lambda_1.t} - e^{-\lambda_0.t}].\lambda_1.dt + w_2.\frac{\lambda_0}{(\lambda_0 - \lambda_2)}.[e^{-\lambda_2.t} - e^{-\lambda_0.t}].\lambda_2.dt$$

with

$$w_2 = 1 - w_1$$

In our expression for $R(t).dt$ the 1/e time constant $\tau_0 = 1/\lambda_0$ controls the signal build-up [5]. We note that in the limit $\lambda_0 \to \infty$ the expression for $R(t).dt$ morphs into the same form as that used for $F(t).dt$, as it should, although in principle one may use different values for λ_A and λ_1 , and λ_B and λ_2 .

Note that the inclusion of $F(t).dt$ in $f(t).dt$ makes $f(t).dt$ finite at time $t = 0$ while the inclusion of $R(t).dt$ allows the signal to also build-up at later times before the entire trend is for decay.

DOUBLES GATE UTILIZATION FACTOR

The Doubles STI GUF, f_d, represents the fraction of Doubles events on the pulse train that fall into the finite STI shift register coincidence gate. The prescription is given by Croft et al [1]:

$$f_d = 2.\int_0^{\infty} f(t).\left[\int_{t+T_p}^{t+T_p+T_g} f(t').dt'\right].dt$$

In this expression T_p is the predelay (the time between the trigger event and the opening of the gate) and T_g the coincidence gate width which together define the early gate structure of the shift register analysis. Evaluation of the expression for f_d may be accomplished using standard forms. The result is:

$$f_d = \alpha^2.f_{Fd} + 2.\alpha.\beta.(I_{FR} + I_{RF}) + \beta^2.f_{Rd}$$

where f_{Fd} is the Doubles GUF for distribution $F(t)$ and f_{Rd} is the Doubles GUF for the distribution $R(t)$. The integrals I_{FR} and I_{RF} represent the cross terms between the functions $F(t)$ and $R(t)$.

$$f_{Fd} = \left(w_A^2 + 2.w_A.w_B.\frac{\lambda_B}{\lambda_A + \lambda_B}\right).f_A + \left(w_B^2 + 2.w_A.w_B.\frac{\lambda_A}{\lambda_A + \lambda_B}\right).f_B$$

$$f_{Rd} = 2.w_1^2.\frac{\lambda_0^2.\lambda_1}{(\lambda_o - \lambda_1)^2}.\left[\frac{1}{2.\lambda_1} - \frac{1}{\lambda_o + \lambda_1}\right].f_1 + 2.w_1.w_2.\frac{\lambda_0^2.\lambda_2}{(\lambda_0 - \lambda_1).(\lambda_o - \lambda_2)}.\left[\frac{1}{\lambda_1 + \lambda_2} - \frac{1}{\lambda_o + \lambda_1}\right].f_1 - 2.w_1^2.\frac{\lambda_o.\lambda_1^2}{(\lambda_o - \lambda_1)^2}.\left[\frac{1}{\lambda_o + \lambda_1} - \frac{1}{2.\lambda_o}\right].f_o$$

$$I_{PR} = w_A.\lambda_A.w_1.\lambda_0/(\lambda_0 - \lambda_1).f_1.1/(\lambda_A + \lambda_1) - w_A.\lambda_A.w_1.\lambda_1/(\lambda_0 - \lambda_1).f_0.1/(\lambda_A + \lambda_0) + w_A.\lambda_A.w_2.\lambda_0/(\lambda_0 - \lambda_2).f_2.1/(\lambda_A + \lambda_2) - w_A.\lambda_A.w_2.\lambda_2/($$

λ_0

where we have the short hand notation:

$f_x = e^{-\lambda_x T_p}.\left(1 - e^{-\lambda_x T_g}\right)$, for x ={A, B, 0, 1, 2}.

which we recognize as being the Doubles STI GUF for a pure exponential decay profile with 1/e time constant $\tau_x=1/\lambda_x$. The expression for f_{Rd} is symmetric to the exchange of subscripts 1 to 2 and 2 to 1 and in the limit $\lambda_o \rightarrow \infty$ the form of f_{Rd} simplifies to that of f_{Pd} as it should [1] for the case of instantaneous build-up. It is important to note that the expression for the f_d is not just a simple linear combination of f_{Pd} and f_{Rd} because of the presence of the cross terms.

THE RANDOM TRIGGERED INTERROGATION GUF FOR DOUBLES

The RTI GUF, g_d is needed when one wishes to make *á priori* estimates of precision on the Doubles rate [3]. It also occurs in expressions for the Triples rate and for the Doubles rate when random (or periodic) triggering of the coincidence gate is used [6]. To compute g_d from f(t) we use [1]:

$$g_d = \int_0^{T_g} \frac{dx}{T_g}.\left[\int_0^{T_g - x} f(t).dt\right]$$

Evaluation of the integral is more straightforward than for the case of f_d and yields the following result, which is a simple linear combination:

$g_d = \alpha.g_{Pd} + \beta.g_{Rd}$

$$g_{Pd} = 1 - w_A.\frac{\left(1 - e^{-\lambda_A.T_g}\right)}{\lambda_A.T_g} - w_B.\frac{\left(1 - e^{-\lambda_B.T_g}\right)}{\lambda_B.T_g}$$

$-\lambda_0$

For finite T_g (and other quantities) in the limit $\lambda_o \rightarrow \infty$, corresponding to instantaneous build-up ($\tau_0=1/\lambda_0=0$) the expression for g_{Rd} takes on the same mathematical form of g_{Pd} as expected.

APPLICATIONS

If the capture time distribution can be measured, or, in the case of a design study simulated (e.g. by Monte Carlo computation) then our analytical expression for $f(t).dt$ gives a convenient means to represent the shape for subsequent calculations. An example would be computing GUFs as a function of predelay and gate width, in order to extract rates over different time intervals. Both gate factors are needed to generate *á priori* estimates of the precision and the optimum gate width settings, which gives the lowest relative precision. The optimum settings are strictly item specific [5] since, as already mentioned, σ_D depends on both temporal behavior and rates both of which may be item specific. However, to simplify calibration and because the optimum sits in a shallow valley usually the same gate settings are used for all items.

Another way of extracting the parameters for the $f(t).dt$ function is to use the expression for STI Doubles GUF derived from it to fit experimental Doubles data taken on a fixed item as either as a function of predelay or as a function of gate width. This makes use of the relationship:

$$D = D_o . f_d \left(T_p, T_g, w_A, \lambda_A, \lambda_B, \lambda_o, w_1, \lambda_1, \lambda_2\right)$$

which rests on the fact that for a given measurement item the observed net Doubles rate varies in direct proportion with f_d. In this expression D_o is the Doubles rate for perfect gating ($T_p = 0, T_g = \infty$) when the function $f_d() = 1$.

In Figure 1 we present an example of a fit to Monte Carlo simulated data. The data represents the capture time distribution of prompt spontaneous fission ^{244}Cm neutron events recorded in the ^{3}He detectors of a particular detector design. The data were generated using the MCNPX code [7] via post processing of the new PTRAC (Particle Track) capture output file. The MCNPX F8 coincidence capture tally allows Doubles rates with perfect and finite gating to be generated and hence f_d values to be extracted. However it does not provide g_d and so the method developed here is needed. The measurement configuration simulated is a prototypical Differential Die-Away Self-Interrogation (DDSI) nondestructive assay instrument surrounding a spent pressurized water reactor fuel assembly underwater. Details of the detector design and this application may be found elsewhere [8]. The fuel assembly, a Westinghouse 17x17 pin design having characteristics typical of discharge from a commercial power plant. In fact the shape is a composite of eleven fuel assemblies with enrichments of 2, 3, 4 and 5 wt% and burn-ups of 15, 30, 45 and 60 GWd/tU, all 5 year cooled, since it was found the shape was closely similar in all cases (the five assemblies of these 16 combinations with negative end of life reactivity were excluded in the present analysis as not of practical interest – that is only the 4 and 5 wt% enrichment were allowed to burn to 45GWd/tU and only the 5wt% fuel allowed to reach 60GWd/tU). It is evident that the proposed functional form is able to reproduce the prompt capture time distribution reasonably well.

In Figure 2 we show a similar plot but this time for events coming from induced fission in the principal fissile constituents namely ^{235}U, ^{239}Pu and ^{241}Pu. In this case the shape changes across the eleven fuel assemblies, for example the time to peak shifts, but for illustrative purposes we again show the composite. Again we find that the proposed returning shape can represent the distribution. The value of S in the expression for $f(t).dt$ is non-zero because the MCNPX data has been normalized to unity over the 0-200 µs interval (rather than over all time).

Figure 3 illustrate s the complete time behavior from a spent fuel assembly. In this case the prompt and returning components have equal contribution which corresponds to a multiplication factor of $M = 2$. The overall shape of the fit to the sum is once again reproduced to a degree considered adequate so that at least the RTI and STI GUF may be estimated for scoping purposes (in use they would be part of the calibration factors). The results for the three profiles are tabulated below.

Trace	f_d	g_d
P	0.772	0.661
R	0.641	0.282
f	0.679	0.465

Table I. Gate Utilization Factors for the Three Traces P, R and f Shown in Figures 1, 2 and 3 Respectively for a Predelay of 4.5 μs and a Gate Width of 64 μs.

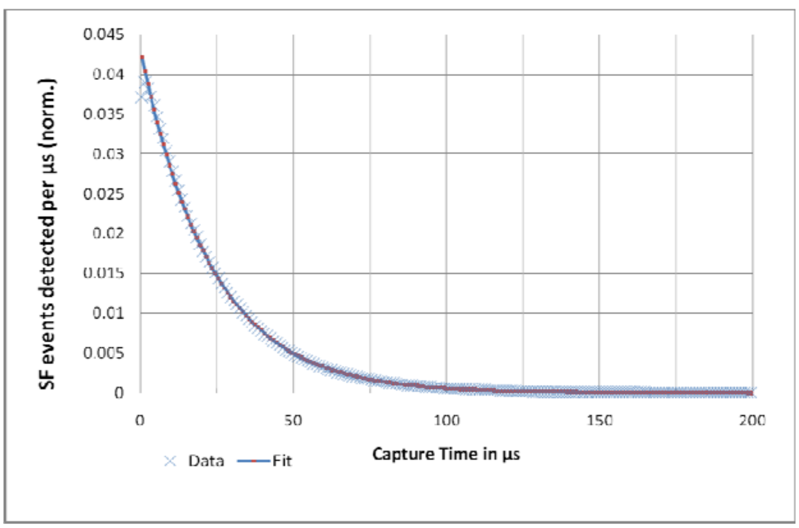

Figure 1. Fit of P(t) to simulated data over the capture time range 5 to 100 μs after the trigger event with the following model parameters: Normalization, S=1.00, w_A=1.0, λ_A=1/23.2 μs^{-1}.

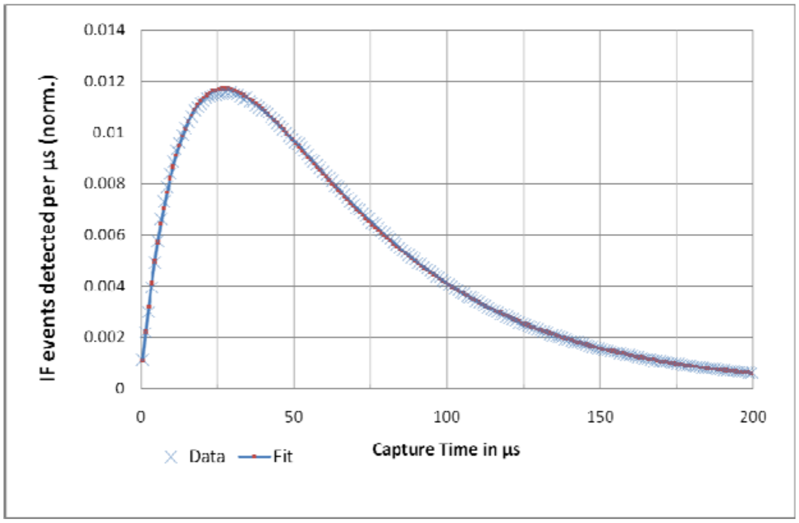

Figure 2. Fit of R(t) to simulated data over the capture time range 0 to 200 µs after the trigger event with the following parameters: Normalization, S=1.03, λ_o=1/16.4 μs^{-1}, w_1=0.0085, λ_1=1/0.0612 μs^{-1}, λ_1=1/51.5 μs^{-1}.

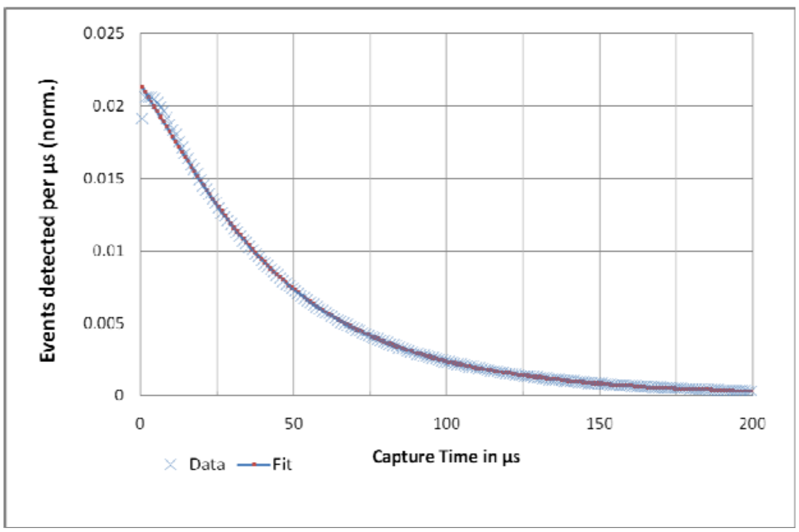

Figure 3. Fit to f(t) shown over the capture time range 0 to 200 µs after the trigger event for a simulated 50:50 blend of the prompt and returning signatures corresponding to a leakage multiplication factor of M=2. S=1.016, α=0.482 with the temporal parameters fixed as per the individual contributions.

DISCUSSION

In selecting values for the predelay, T_p, and gate width, T_g, it is usual to minimize the relative statistical counting precision on the Doubles rate. The best choice of predelay is the shortest setting which does not introduce an instrumental bias. For a simple exponential dieaway profile the optimum gate width setting is commensurate with the 1/e dieaway time, about 1.2 times the dieaway time is a traditional rule of thumb [4]. For the present problem the time behavior is more complex and the best choice of gate must be picked with care. We note that with finite λ_o the Doubles lost to the predelay are lessened. Also for the model presented the optimum gate width is in general shifted to higher values than $\sim 1.2 . \tau_A$. In addition we recognize and intend to exploit the fact that important information about the fissile content of the fuel can be extracted from the time trace including using various gate structures to preferentially emphasize the prompt and returning components. Since the results are counter and item specific it is difficult to make general statements. However, when the counter efficiency is modest (and for spent fuel an efficiency of even 1% will challenge current count rate capability at the higher fuel burnups yet be adequate for Doubles counting) the optimal gate width when the Accidentals rate far exceeds the Doubles rate is obtained by minimizing $\sqrt{T_g}/f_d$. Using this criterion we find that for the three time traces considered in this article (P, R and f from Figures 1, 2 and 3 respectively) that for a predelay of 4.5 μs the gate width that minimizes the relative counting precision on the overall Doubles is about 29.1, 73.4 and 55.2 μs respectively. The best gate width values to be used when analyzing composite time traces with an eye to teasing apart the two temporal contributions is beyond the remit of the present paper.

We observe that in Figure 1 even the prompt component appears to exhibit a build-up behavior but on a much shorter time scale. Also the dieaway is rapid so that by 100 μs most of the intensity is included. Consequently in our fitting, which was based on minimizing the sum of fractional differences between the MCNPX trend line and the fitted trend line at the centroid of each 1 μs wide bin, we restricted the range of the fit to the interval 5 to 100 μs and obtained a good fit with just a single time constant. However, formally we could have easily used the functional form including build-up and since the result for g_d is just a simple linear combination we could have achieved a slightly more refined estimate for its value in a straight forward fashion. However, since our primary goal was to extract g_d for use in precision estimates, which according to the semi-empirical formula being applied [3] are perhaps good to only 10-20% the approach described is considered sufficient.

CONCLUSIONS

Advanced ^{3}He-based neutron correlation counters are being considered for the assay of spent fuel assemblies in order to quantify fissile mass. Spent fuel assemblies are highly multiplying and exhibit long fission chains in part due to (thermal) neutrons which return from the surroundings later in time than the spontaneous fission events which initiate the chains. This results in a capture time distribution which is quite different to the monotonic decay previously considered until the present effort. In this work we have proposed a new fitting function and showed how it can be used to fit & smooth Doubles data and to estimate Signal Triggered Interrogation Doubles Gate Utilization Factors f_d and Random Triggered Interrogation Gate Utilization Factors g_d respectively. The latter is used in *á priori* performance estimates but is not directly available from transport codes.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the support of the Next Generation Safeguards Initiative (NGSI), funded through the U.S. Department of Energy's National Nuclear Security Administration's Office of Dismantlement and Transparency (NA-241).

REFERENCES

1. CROFT S., MCELROY R.D. and KANE S.C. (2007) Coincidence gate utilization factors for neutron correlation counters with up to three components in the die-away profile, Proceedings of the ICEM '07: The 11th International Conference on Environmental Remediation and Radioactive Waste Management, Bruges (Brugge), Belgium, 2-6 September 2007. Copyright ©2007 by ASME, ISBN 0-7918-3818-8 (2007). Paper 7173.
2. BÖHNEL K. (1978) Determination of plutonium in nuclear fuels using the neutron coincidence method, Kernforschungszentrum, Karlsruhe Report KFK 2203, AWRE, Aldermaston, Berks., UK, Translation No. 70(54/4252). ISBN-0-85518121-4(Mar. 1978).
3. DYTLEWSKI N., KRICK M.S. and ENSSLIN N. (1993) Measurement variances in thermal neutron coincidence counting, Nucl. Instrum. & Meths. in Phys. Res. A327(1993)469-479.
4. CROFT S., EVANS L.G. and SCHEAR M. (2010) Optimal gate-width setting for passive neutron multiplicity counting, 51st Annual Meeting of the Institute of Nuclear Materials Management, July 11-15, 2010, Baltimore, MD, USA.
5. CROFT S., SCHEAR M. and EVANS L.G. (2010) Analytical expression for the gate utilization factors of passive multiplicity counters including signal build-up, 51st Annual Meeting of the Institute of Nuclear Materials Management, July 11-15, 2010, Baltimore, MD, USA.
6. CROFT S., MCELROY R.D., PHILIPS S., VILLANI M.F. and EVANS L.G. (2007) Dead Time Behaviors in Passive Neutron Correlation Counting, Waste Management Symposia, WM'07 February 25 – March 1 2007, Tucson, Arizona, USA. Paper 7258.
7. PELOWITZ J.F., et. al. (2005) MCNPX User's Manual Version 2.5.0, Los Alamos National Laboratory report, LA-CP-05-0369(2005).
8. SCHEAR M.A., MENLOVE H.O., TOBIN S.J. and EVANS L.G. (2010) Fissile Material Measurements using the Differential Die-Away Self-Interrogation Technique, Institute of Nuclear Materials Management 51st Annual Meeting, Baltimore, Maryland (2010).