Annals of Nuclear Energy 99 (2017) 36–39

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Incorporating delayed neutrons into the point-model equations routinely used for neutron coincidence counting in nuclear safeguards

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ARTICLE INFO

Article history:
Received 20 May 2016
Received in revised form 15 August 2016
Accepted 19 August 2016

Keywords:
Delayed neutrons
Neutron coincidence counting
Point-model equations
Neutron multiplicity counting

ABSTRACT

We extend the familiar Böhnel point-model equations, which are routinely used to interpret neutron coincidence counting rates, by including the contribution of delayed neutrons. After developing the necessary equations we use them to show, by providing some numerical results, what the quantitative impact of neglecting delayed neutrons is across the full range of practical nuclear safeguards applications. The influence of delayed neutrons is predicted to be small for the types of deeply sub-critical assay problems which concern the nuclear safeguards community, smaller than uncertainties arising from other factors. This is most clearly demonstrated by considering the change in the effective (α,n)-to-spontaneous fission prompt-neutron ratio that the inclusion of delayed neutrons gives rise to. That the influence of delayed neutrons is small is fortunate, and our results justify the long standing practice of simply neglecting them in the analysis of field measurements.

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1. Introduction

Neutron coincidence counting (NCC) is a well-established and widely used technique for non-destructively assaying the plutonium content of items (Ensslin, 1991; Ensslin et al., 1998; Pázsit et al., 2009). The conventional target quantity of the assay is the ^{240}Pu-effective mass, a weighted linear sum of ^{238}Pu, ^{240}Pu and ^{242}Pu. The total plutonium mass can then be derived from the ^{240}Pu-effective mass from independent knowledge of the isotopic composition of the plutonium. The primary measured quantities in NCC are the singles and doubles rates. The singles count rate, S, is the total neutron time-averaged event rate on the pulse train and is often referred to as the totals rate. The doubles (reals, pairs or coincidence) rate, D, is most often determined using a form of autocorrelation analysis known as shift-register logic (Ensslin, 1991). For practical non-destructive assay applications the two rates are expressed in terms of a combination of item and detector parameters, and nuclear data constants, using algebraic expressions based on a one-group, point, prompt-kinetics model (Bohnel, 1985; Favalli et al., 2015; Cifarelli and Hage, 1986; Croft et al., 2012, 2015a,b, 2016; Croft and Favalli, 2012) – the so called Böhnel point-model equations (Ensslin, 1991; Ensslin et al., 1998; Pázsit et al., 2009; Bohnel, 1985). These can then be solved for the ^{240}Pu-effective mass. The standard formulation of the point-model equations neglects delayed neutrons, this is an approximation. In this article we quantify what impact this routinely applied approximation has on assay results across the domain of nuclear safeguards.

Our starting point is the familiar point-model equations for the singles and doubles counting rates. These may be expressed as follows:

$$S = F_S \varepsilon M_L (1+\alpha) \nu_{S1} \quad (1)$$

$$D = F_S \varepsilon^2 f_d M_L^2 \frac{\nu_{S2}}{2}\left[1 + \frac{\nu_{S1}(1+\alpha)}{\nu_{S2}/2}\left(\frac{M_L - 1}{\nu_{I1} - 1}\right)\frac{\nu_{I2}}{2}\right] \quad (2)$$

where the variables used are defined as follows:

F_S = the effective ^{240}Pu spontaneous fission rate = $m_{eff} \cdot g$, the product of the ^{240}Pu-effective mass, m_{eff}, and g, the specific spontaneous fission rate of ^{240}Pu,

ε = the neutron detection efficiency (probability of detection per neutron emerging from the item),

ν_{Si} = ith factorial moment of the spontaneous fission neutron distribution,

ν_{Ii} = ith factorial moment of the induced fission neutron distribution,

M_L is the neutron leakage self-multiplication $M_L = M_T \cdot p_L = (1-p_f-p_c)/(1-\nu_{I1}p_f)$ where $M_T = 1/(1-\nu_{I1}p_f)$ is the total self-multiplication, $p_L = 1 - p_f - p_c$ is the probability of neutron

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http://dx.doi.org/10.1016/j.anucene.2016.08.012
0306-4549/Published by Elsevier Ltd.

leakage, where p_f and p_c are the probabilities for a neutron to undergo fission and parasitic captures (Croft et al., 2012, 2015b; Pázsit, 2016),

α = the (α,n)-to-spontaneous fission prompt-neutron production ratio, i.e. (α,*n*)-*rate*/($F_S \cdot \nu_{S1}$),

f_d = the doubles gate utilization factor (Ensslin et al., 1998; Croft and Favalli, 2012; Henzlova et al., 2015).

For the present discussion we imagine the common application in which the properties of the detector (efficiency and gate utilization factor) are assumed to be known from calibration and the α-value can be calculated from the isotopic composition of the α-emitting constituents of the measurement items (e.g. ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am) and chemical form (e.g. dioxide) of the chemically pure compound inside the item.

2. Derivation of Böhnel-like point model equation with delayed neutrons embedded

2.1. Delayed neutrons and assumptions in the model

Passive neutron coincidence counting assays are performed in steady state conditions when the neutron emission processes have settled into a quiescent dynamic equilibrium. Thus we can speak in terms of the time averaged behavior. Delayed neutrons are emitted long after the associated prompt fission neutrons (which for our discussion are emitted essentially instantaneously at the time of fission), on a typical time scale of $\sim$1/10–100 s (Keepin et al., 1957; Walker and Weaver, 1979). This is also much longer than the time prompt fission neutrons take to emerge from measurement item and subsequently persist in the detector system (for commonly used moderated ^{3}He based detection systems the characteristic detector die-away time is of the order of 10's μs) (Ensslin, 1991; Croft et al., 2012, 2016). Thus, as a practical matter, the fundamental assumption we introduce in our treatment is that DN's have the essential characteristics of time random neutrons, like (α,n) neutrons, being created one at a time and independent in time from the parent fission events (see also Fermi, 1942; Uhrig, 1970 for discussion of delayed neutrons in a reactor close to critical) This is a key assumption in order to introduce DN contributions into the Böhnel-like point model equations, as we will do in the next chapter. For the discussion, it is also worth noting that, in the Böhnel point-model equations, time never appears explicitly. One derives first the distribution of the total number of neutrons leaving the sample per one starting neutron, and after that per one source event (spontaneous fission neutrons with a number distribution), assuming that all these neutrons (including internal multiplication before leakage) are all generated "instantaneously" (the idea of "superfission" introduced by Bohnel (1985). Time, in the form of count rates of singles, doubles etc., which means detection per unit time, enters only by assuming a source intensity for the spontaneous fissions (and of course (α,*n*) neutrons), but assuming that the fission chains do not overlap (the concept of superfission guarantees this, since the evolution of the chains takes zero time). Hence the doublets, triplets etc. are calculated from the generation rate of the multiplets and the detection probability of neutrons. Finite (between zero and unity) gate factors account for the use of finite coincidence gates and the finite residence time of neutrons in the detector. Delayed neutrons, however, by definition, cannot be accommodated into the concept of superfission. But our assumption that all neutrons which originate from a delayed neutron precursor represent only time-uncorrelated 'background' allows us to introduce the DN contribution in the Böhnel point-model equations like a special kind of (α,n) contribution. By this assumption we avoid having to deal explicitly with connected time-dependent processes, because neutrons belonging to the prompt-fission chain are separated from those generated by delayed neutron precursors. The former gives the correlated counts, the latter the corresponding uncorrelated background. A full derivation by master neutron stochastic equation is in preparation (Pázsit), but it goes beyond the scope of this paper, which is ultimately to quantify the contribution of the delayed neutron production in the Böhnel point-model equations and applied in the nuclear safeguards and also waste management assay scenarios.

2.2. Derivation of the equations

Each spontaneous fission (SF) gives rise to an average of ν_{Sd} DN's, and each induced fission (IF) gives rise to an average of ν_{Id} DN's. In the one neutron energy-group point-model (Croft et al., 2012), each neutron initially released inside the measurement item also results, again on average, in a number of IF's given by:

$$p_f \cdot M_T = \frac{M_T - 1}{\nu_{I1}} \approx \left(\frac{M_L - 1}{\nu_{I1} - 1}\right), \text{ IF per n} \tag{3}$$

where:

p_f is the probability that the initial neutron history will end in absorption resulting in fission;

M_T is the total self-multiplication = $1/(1-\nu_{I1} p_f)$;

and, the approximate form on the right hand side holds when the probability of parasitic p_c (e.g. (n,γ)) neutron capture in the item relative to the corresponding probability of induced fission p_f is small.

In nuclear safeguards of plutonium, the items of interest are usually deeply sub-critical (Ensslin, 1991; Ensslin et al., 1998), and, we are concerned with quantifying the mass of Pu present through the ^{240}Pu-effective mass which is used as a measure of the overall SF rate. We shall therefore treat the SF process as the primary initiating event and scale primary ('source term') neutron production from it. Now, on the average F_S SF's take place per second and these in turn liberate ($F_S \cdot \nu_{S1}$) prompt neutrons per second into the system along with ($F_S \cdot \nu_{Sd}$) delayed neutrons, and [($F_S \cdot \nu_{S1}$)$\cdot\alpha$] (α,n) neutrons. Thus, the rate at which primary-source neutrons are released into the measurement item is given by:

$$F_S \nu_{S1} \left(1 + \alpha + \frac{\nu_{Sd}}{\nu_{S1}}\right) \tag{4}$$

and hence the rate of IF DN production is obtained by the product of Eq. (4), Eq. (3), and the IF DN yield as follows:

$$F_S \nu_{S1} \left(1 + \alpha + \frac{\nu_{Sd}}{\nu_{S1}}\right)\left(\frac{M_L - 1}{\nu_{I1} - 1}\right)\nu_{Id} \tag{5}$$

The IF DN contribution given by Eq. (5), like the SF DN's, have the essential (mathematical) characteristics of additional (α,n) events. Thus, in the standard point-model equations, Eqs. (1) and (2), we should replace the term (1 + α) by the following term:

$$\left(1 + \alpha + \frac{\nu_{Sd}}{\nu_{S1}}\right)\left(1 + \left(\frac{M_L - 1}{\nu_{I1} - 1}\right)\nu_{Id}\right) \tag{6}$$

For non-multiplying items the second factor in brackets reduces to zero. For chemically pure metallic items we would expect (α,n) production to be close to zero and so, the time-random neutron production will be influenced most strongly by the DN component for such materials. This may be the case for certain calibration items, for example.