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July 1, 2016

SORMA West 2016  
Berkeley, CA, United States  
May 23, 2016 through May 26, 2016

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# Characterization of fissile assemblies using low efficiency detection systems

George F. Chapline and Jerome M. Verbeke

*Abstract*— Determining the amount, chemical form, and physical shape of the fissile material in an unknown assembly is a challenging problem that is of great importance in national security contexts. Unfortunately, current methods (e.g., measurements of neutron correlations with He3 detectors) typically require a detection system with a geometric efficiency that would be impractical to realize in the field. In addition current methods are challenged when the  $\alpha$ -ratio of the SNM is completely unknown because the equations become non-deterministic if the  $\alpha$ -ratio is unknown [1]. In this paper we show that timely characterizations of fissile material can be obtained using fast neutron/ $\gamma$ -ray detection systems whose efficiency is too low for current passive characterization methods to be useful. Our method relies on combining measurements of the spectrum of correlated fast neutrons with an analytic relationship between the positions of the peaks and minima in a plot of the distribution of time intervals  $\Delta t$  between neutrons/ $\gamma$ -rays as a function of  $\log \Delta t$  and the amount and multiplication of the fissile material. These positions depend only weakly on detector efficiency, and this dependence is only via the observed count rate; i.e. the detector efficiency doesn't have to be explicitly known. A crucial ingredient for our new method is our discovery that whether an unknown fissile material is in a metallic or oxide form can be very rapidly resolved by comparing the spectra of correlated and uncorrelated neutrons. We illustrate our method using Pu and HEU samples.

*Index Terms*— Correlated neutrons, fast neutron counting, scintillator detectors.

## I. INTRODUCTION

Characterizing the fissile material in an object of unknown composition typically requires a neutron/ $\gamma$ -ray detection system with good geometric efficiency. We have been investigating the possibility that timely estimates of the amount, form, and multiplication of the fissile material in an assembly can be obtained using relatively low efficiency detection systems by observing the positions of the peaks and minima in a plot of the distribution of time intervals  $\Delta t$  between fast neutrons as a function of  $\log \Delta t$ . It is noteworthy in this connection that the position of the correlated neutron peak is practically independent of detector efficiency, while the position of the first minimum between the correlated and uncorrelated peaks is only weakly dependent on efficiency. Furthermore this weak dependence can be approximately calculated using only the observed count rate. This is an enormous improvement over current characterization methods (e.g. the Cifarelli-Hage method [1]) which require that the detector efficiency be explicitly determined.

Our new approach is based on the realization that the positions of the peak of the interval time distribution for correlated neutrons, and the minimum the interval time distribution in a plot of the distribution of time intervals  $\Delta t$  between pairs of fast neutrons as a function of  $\log \Delta t$  (see Fig. 1) depends only weakly on the detector efficiency. We have discovered in experiments using liquid scintillators that observation of these characteristic interval times can be combined with comparison of the fast neutron spectra for correlated and uncorrelated neutrons (see Fig. 2) that one can rapidly determine whether the fissile material is in a metallic or oxide form, estimate the amount of fissile material, and determine whether it is shaped as a ball or a shell. In section II we describe the analytical theory that allows us to relate the maximum and minimum of the interval time distribution for time correlated neutrons on the left side of Fig. 1 to the amount and form of fissile material.

Of course a scintillator detector cannot directly measure the spectrum of neutron energies because the scintillator only measures the amount of light produced by protons recoiling after being hit by a neutron. However we have discovered that the differences in

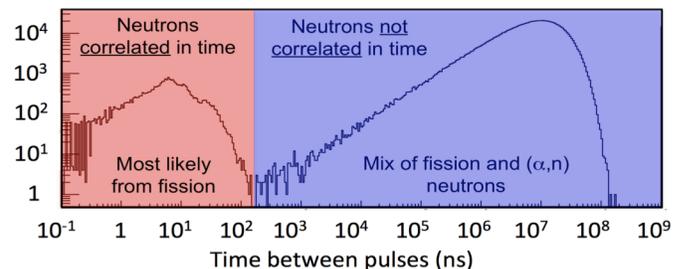


Fig. 1. Typical distribution of interval times between fast neutrons for materials containing U or Pu measured with liquid scintillator detectors.

This paper was submitted for review on July 1, 2016. This work was supported in part by the U.S. Department of Energy under Contract DE-AC52-07NA27344. G. F. Chapline is with the Physical and Life Sciences Directorate, Lawrence Livermore National Laboratory, Livermore, CA 94551 USA. (e-mail: chapline1@llnl.gov).

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the distribution of pulse heights observed in a liquid scintillator for fission neutrons and ( $\alpha$ ,n) neutrons can be clearly distinguished experimentally. This difference allows us to immediately distinguish whether the fissile material is in an oxide or metallic form. In addition in section III we describe how liquid scintillators can in some cases be used to quantitatively determine the  $\alpha$ -ratio; i.e. the relative contribution of spontaneous fission and ( $\alpha$ ,n) neutron emission to the rate of neutron emission. In section IV we compare our estimates for the amount and multiplication of a metallic Pu ball and small sample of PuO<sub>2</sub>

## II. ANALYTIC THEORY OF INTERVAL TIMES DISTRIBUTION

Prasad et al. [2] have shown that the statistical distribution  $I(\Delta t)$  of time intervals between two neutron counts can be obtained from the formulae

$$I(\Delta t) = \frac{d}{d\Delta t} [1 - n_0(\Delta t)] \quad (1)$$

$$n_0(\Delta t) = \frac{1}{R_1} \frac{d}{d\Delta t} [1 - b_0(\Delta t)] \quad (2)$$

where  $n_0$  is the probability of no counts in a time interval  $\Delta t$  following a trigger neutron count and  $R_1$  is the average neutron count rate. It can be shown [2] that when the fissile multiplication is not large  $b_0$  can be calculated from the Feynman-de Hoffman 2-neutron correlation  $Y_{2F}(\Delta t)$  [3]:

$$b_0(\Delta t) = \exp[-R_1 \Delta t (1 - Y_{2F}(\Delta t))] \quad (3)$$

where

$$Y_{2F}(\Delta t) = R_{2F}(M, A) \left( 1 - \frac{1 - e^{-\alpha \Delta t}}{\alpha \Delta t} \right) \quad (4)$$

The constant  $\alpha$  represents the relaxation rate for the number of neutrons inside the fissile assembly, while  $R_{2F}$  is a constant that depends on the average number of neutron pairs emitted per fission, the multiplication  $M$  and  $\alpha$ -ratio  $A$  [3]. Experimentally this constant represents the ratio of the areas under the 2 peaks in Fig. 1 Substituting the expressions (2) -(4) into (1) yields the following analytic expression for the interval time distribution:

$$I(\Delta t) \cong \exp[-R_1 T (1 - Y_{2F}(\Delta t))] \{ R_1 [1 - R_{2F} (1 - e^{-\alpha \Delta t})]^2 + \alpha R_{2F} e^{-\alpha \Delta t} \} \quad (5)$$

Similar formulas have been worked for the interval times between 2  $\gamma$ -rays or a  $\gamma$ -ray and neutron, but these formulae necessarily involve more than one relaxation time. The main point we would to emphasize in this paper is that the time intervals  $\Delta t$  corresponding to peak of the interval time distribution on the left side of Fig.1 representing correlated fast neutrons and the minimum in the middle of the figure between the correlated and uncorrelated peaks in the interval time distribution provide information regarding the amount, chemical form, and shape of the fissile source of neutrons in an assembly that is only weakly dependent of on detector efficiency. The values for these interval times can be found by differentiating with respect to  $\log \Delta t$ :

$$\frac{d}{d\Delta t} (\Delta t I(\Delta t)) = 0 \quad (6)$$

which yields an analytic formula for the maxima and minima of the interval time distribution  $I(\Delta t)$ :

$$\left[ 1 + \frac{\alpha R_{2F}}{R_1} (1 - \alpha \Delta t) e^{-\alpha \Delta t} \right] = R_1 \Delta t \left\{ 1 + \frac{\alpha R_{2F}}{R_1} \left[ e^{-\alpha \Delta t} + \frac{2}{\alpha \Delta t} (1 - (1 - \alpha \Delta t) e^{-\alpha \Delta t}) \right] \right\} \quad (7)$$

The values of  $\Delta t$  which satisfy (7) as a function of  $\alpha R_{2F}/R_1$  are shown in Fig. 2.

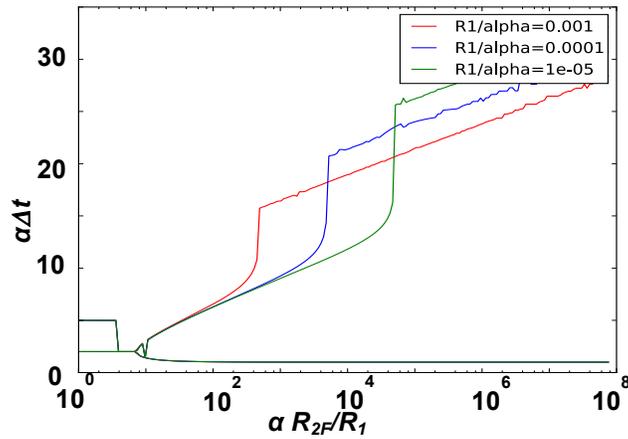


Fig. 2: Solutions to (7) which relate the values of  $\Delta t$  at the peak and minimum in  $I(\Delta t)$  to  $\alpha$  and  $R_{2F}/R_1$

Fig. 2 shows that the position of the peak in the interval time distribution for correlated neutrons is for all practical purposes independent of the neutron count rate, while the minimum in the interval time distribution between the peak of interval time distribution for correlated neutrons and the broad peak in the interval time distribution associated with uncorrelated neutrons depends only weakly on the count rate for neutron emission. Of course, if there are neutrons from other sources, e.g. neutrons generated by cosmic rays, then this separate source of neutrons must be accounted for separately. As is evident from the curves in Fig. 2 the peak of the interval distribution for correlated neutrons equal to the relaxation time  $1/\alpha$  in our single relaxation time approximation. Actually it is only approximately true that the time dependence of the neutron population in a fissile assembly can be described with a single relaxation time  $1/c$ . In reality there will be multiple time scales associated with the time evolution of fission chains and leakage of neutrons from the assembly. Fortunately though we have found that for many situations of practical interest the assumption that there is a single relaxation time leads to a good first guess for the amount and shape of the fissile material.

This weak dependence on detector efficiency is illustrated in Fig. 3, which shows the fast neutron interval time distribution for a 2kg Pu ball, measured with arrays with 77 and 10 liquid scintillator cells. As is evident from Fig. 3 the position of the minimum in  $I(\Delta t)$  depends only weakly on the detector efficiency. This weak dependence of the minimum in the interval time distribution on detector efficiency allows us to use curves like those shown in Fig. 2 to estimate the amount of fissile material and its multiplication

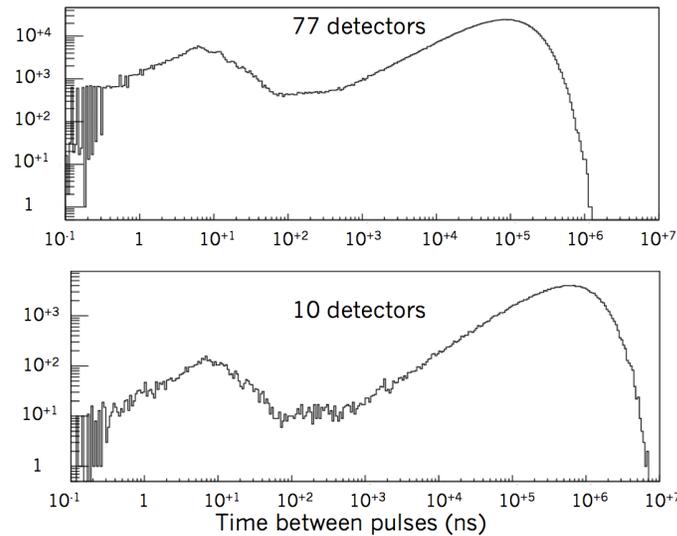


Fig. 3: Effect of reducing the number of detectors in an array of scintillator neutron detectors .

even when the detector efficiency is relatively low. Thus our method contrasts sharply with characterization methods using He3 neutron detectors which typically require relatively good geometric efficiency and explicit knowledge of the detector efficiency. In our approach the only way the detector efficiency enters the analysis is via the observed count rate  $R_1$ . Of course if the count rate  $R_1$  is too low due either to low detector efficiency or the small size of the sample of fissile material, the time needed to construct the interval time distribution may become inconveniently long. In addition, the minimum in  $I(\Delta t)$  is easily seen only if the relaxation rate  $\alpha$  is much faster than the average count rate for uncorrelated neutrons. However we have found that in many situations of practical interest 20 minutes of observing time is sufficient to obtain values of the interval times for the peak and minimum in  $I(\Delta t)$

,which can be combined with an analytic solution to (7) to estimate  $R_{2F}/R_1$ . One can use this estimate for  $R_{2F}/R_1$  and the observed value of  $R_1$  to estimate the amount of fissile material and the multiplication if the  $\alpha$ -ratio is known. In section III we describe how liquid scintillators can also be used to estimate the  $\alpha$ -ratio, which in turn can be used to estimate the multiplication and determine the shape of the fissile material. In section IV we compare our estimates for the amount and multiplication of a metallic Pu ball and small sample of  $\text{PuO}_2$  obtained by combining the observed value of  $R_1$  with values for  $\alpha$  and  $\alpha R_{2F}/R_1$  deduced using the analytic solution to (7) with exact values. Even for the Pu ball with a multiplication near 2 our method yields useful estimates. This is perhaps a little surprising since our method is based on using only the Feynman 2-neutron correlation function to construct the interval time distribution, cf. (3), which strictly speaking is only exact for  $M$  close to 1.

As described in our companion poster [5] these methods also seem to work even if the fissile material is moderated, provided the moderation is not so great that there is no observable emission of fast neutrons. We have found, for example that our method for estimating the amount and form of fissile material does continue to work when the fissile material is moderated with 2" of polyethelene, because even with this amount od moderation one can still construct an interval time distribution for the fast part of the neutron spectrum that resembles Fig. 1.

### III. METAL OR OXIDE?

An essential ingredient in our approach to characterizing an unknown sample of fissile materials is a method for almost immediately determining whether the fissile material is in a metallic or oxide form. Ideally one would like to know the exact  $\alpha$ -ratio, but even if one only knows whether the fissile material is in a metallic or oxide form one make an educated guess as to the  $\alpha$ -ratio, and in addition know what values to assume for the bulk fission time  $\tau_f$ , since this fission time is completely determined by determined by the atomic number of the fissile isotope and its density. Thus knowing whether the fissile isotope is metallic or an oxide can lead one to quickly place a lower bound on the multiplication  $M$  via the point model relation [3]  $M > 1/\alpha\tau_f$ .

Our method for determining whether the fissile material is metallic or an oxide relies on the fact that fast neutrons from fission and  $(\alpha,n)$  processes have different energy spectra (see Fig. 4).

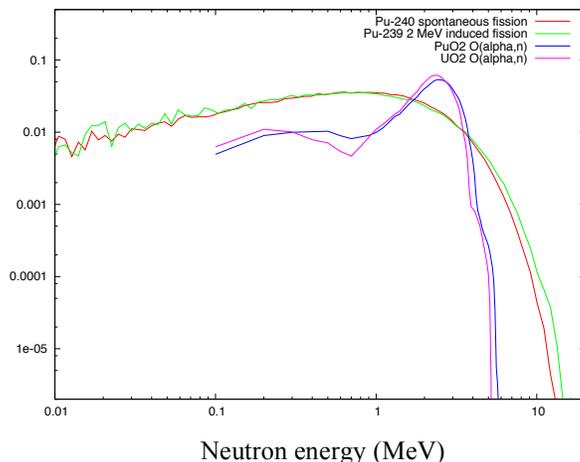


Fig. 4: Energy spectra for fission neutrons and  $(\alpha,n)$  neutrons emitted by Pu and U oxide materials.

We have previously shown [4] that these differences are reflected in the pulse height distribution for fast neutrons seen in a liquid scintillator detector, even though this pulse height distribution measures the energies of recoil protons rather than the neutron energy directly. On the other hand we have previously shown [4] that the different pulse height distributions for fast neutrons from the metallic and oxide forms of fissile materials can by themselves be used to reconstruct the  $\alpha$ -ratio for oxide forms of the fissile material. This is illustrated in Fig. 5 which shows how we were able to determine the  $\alpha$ -ratio for a sample of  $\text{PuO}_2$  by comparing the observed normalized pulsed height distribution with normalized pulse height distributions for Pu fission and HEU oxide. In the case of  $\text{PuO}_2$  the normalized fast neutron pulse height distribution for HEU oxide can serve as a surrogate for the  $(\alpha,n)$  neutron spectrum associated with  $\text{PuO}_2$  because the  $(\alpha,n)$  spectra are similar (cf. Fig. 4) and the  $\alpha$ -ratio for HEU oxide is very large, and therefore the pulse height distribution from HEU oxide is almost entirely due to  $(\alpha,n)$  neutrons.

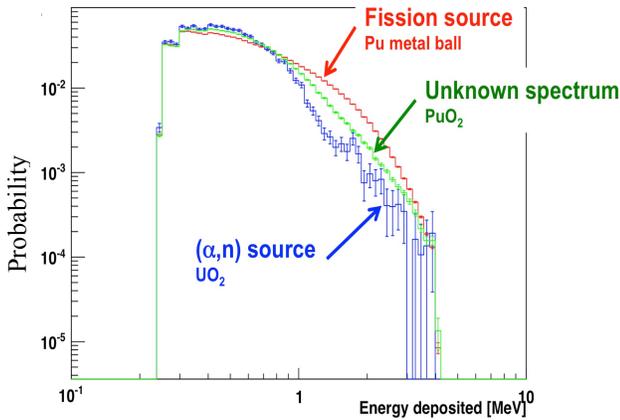


Fig. 5: Reconstruction of liquid scintillator PuO<sub>2</sub> pulse height spectrum from pure fission and (a,n) pulse height spectrum

We have recently shown [5] that when it is not certain what would be a suitable surrogate for the (α,n) neutron spectrum, then it is still possible to almost immediately determine whether the fissile material is in a metallic or oxide form by comparing the pulse height distribution for the correlated and uncorrelated fast neutrons; i.e. for the neutrons associated with the 2 different peaks in the interval time distribution shown in Fig.1. As it happens both the first or second neutrons in a correlated pair are almost certainly fission neutrons. It is theoretically possible that the first neutron is an (α,n) neutron, but it is very unlikely that this (α,n) neutron could scatter in a detector cell and then return to the assembly to cause a fission detected in the scintillator array.

#### IV. TWO EXAMPLES

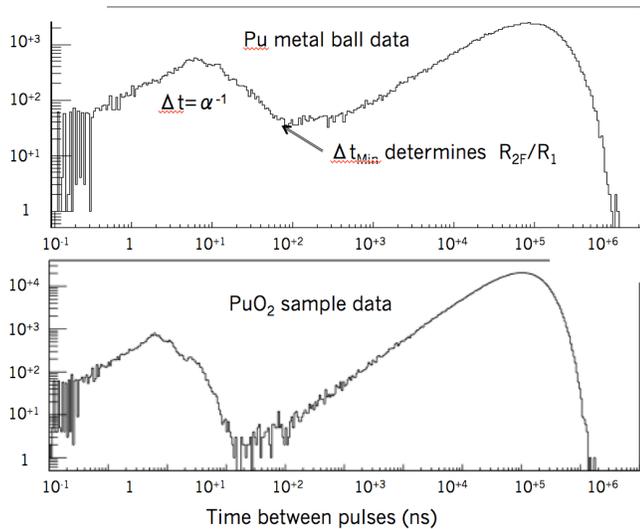


Fig. 6: Fast neutron interval time distribution for a 2.4 kg metallic Pu ball and a 9gm sample of PuO<sub>2</sub>.

In order to illustrate how well our method works for two very different situations: 1) a 2.35 kg metallic ball of Pu with a multiplication near 2 and  $R_1=100\text{Hz}$ , and 2) a 9 gm sample of Pu oxide with  $R_1=10^4\text{ Hz}$ . The maximum of the correlated neutron peak for the metallic Pu ball is very close to the value of  $\alpha^{-1}=6.8\text{ns}$  inferred from Monte Carlo. The minimum in the time interval distribution near 100ns corresponds to  $R_{2F} = 0.26$ , which is larger than the exact value 0.21 (due to neglect of 3-neutron correlations). This leads to an underestimation of the amount of Pu (2.35kg) by 15% and an overestimation of the multiplication by 10%. For the 8.7 gm PuO<sub>2</sub> ( $A = 0.9$ ) sample the maximum of the correlated neutron peak is 6ns, which is significantly smaller than the relaxation time (8.3 ns) obtained from a moment analysis, and leads to a large error in the predicted PuO<sub>2</sub> mass. However, in this case the correlated part of  $I(\Delta t)$  has multiple peaks. If we identify the middle peak with  $\alpha^{-1}$  then the minimum near to 30ns corresponds to  $R_{2F} \approx 0.3$ ; i.e. close to the Monte Carlo prediction 0.28.

#### V. CONCLUSION

We have outlined a new method for using fast neutron detectors to passively characterize the fissile material in an assembly of unknown composition. We have found that our algorithm can be used to obtain useful estimates for the amount and form of fissile material; even when the detector efficiency is too low to permit use of the methods that are typically used with He3 counting methods. In the future we would like to introduce, in addition to the single relaxation rate  $\alpha$  used in the Feynman-de Hoffmann formula, relaxation times corresponding to neutron and  $\gamma$ -ray times of flight. We expect that an improved version of

our algorithm that takes into explicit account time of flight time scales for neutrons and  $\gamma$ -rays will allow us to make better predictions for the size and shape of fissile materials when the sample size is small.

#### ACKNOWLEDGMENT

The authors would like to thank Neal Snyderman and Les Nakae for helpful discussions.

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**Jérôme Verbeke** joined the radiation technology group at Lawrence Livermore National Laboratory in April 2004, where he works on Monte-Carlo codes and authentication and detection of fissile material. Prior to that, he worked four years in the high-performance computing group at Sun Microsystems, Inc., where he was a consultant to the electronic design automation software industry, optimizing third-party scientific and technical applications. Between 1995 and 2000, he developed DD/DT neutron sources and neutron moderators at Lawrence Berkeley National Laboratory. Monte-Carlo codes for radiation transport have been his interest and specialty since 1995. He has published over 50 articles in peer-reviewed technical journals and conferences, and holds 5 U.S. patents. He obtained a Masters degree in Mechanical Engineering from the University of Louvain in Belgium, and a PhD in Nuclear Engineering from the University of California, Berkeley, California, in 2000.