Application of Bayes' theorem for pulse shape discrimination

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Abstract

A Bayesian approach is proposed for pulse shape discrimination of photons and neutrons in liquid organic scintillators. Instead of drawing a decision boundary, each pulse is assigned a photon or neutron confidence probability. This allows for photon and neutron classification on an event-by-event basis. The sum of those confidence probabilities is used to estimate the number of photon and neutron instances in the data. An iterative scheme, similar to an expectation-maximization algorithm for Gaussian mixtures, is used to infer the ratio of photons-to-neutrons in each measurement. Therefore, the probability space adapts to data with varying photon-to-neutron ratios. A time-correlated measurement of Am-Be and separate measurements of ^{137}Cs , ^{60}Co and ^{232}Th photon sources were used to construct libraries of neutrons and photons. These libraries were then used to produce synthetic data sets with varying ratios of photons-to-neutrons. Probability weighted method that we implemented was found to maintain neutron acceptance rate of up to 90% up to photon-to-neutron ratio of 2000, and performed 9% better than decision boundary approach. Furthermore, the iterative approach appropriately changed the probability space with an increasing number of photons which kept the neutron population estimate from unrealistically increasing.

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Keywords: Pulse shape discrimination, Liquid scintillator, Bayes' theorem, Expectation-Maximization

1. Introduction

1.1. Overview of the PSD problem

In organic scintillators the fraction of light emitted during delayed fluorescence depends on the exciting particle, therefore it is possible to differentiate between neutron and photon interactions through pulse shape discrimination (PSD) [1]. Although this property has been known for decades, recent advancements in pulse digitization [2] and the demand for an alternative to the He-3 neutron detectors for security applications [3] have invigorated the interest in PSD performance in organic scintillators. Reliable and robust PSD methods are necessary with organic scintillators to match the gamma rejection capabilities of He-3 detectors.

The charge integration PSD method, in both analog and digital applications, relies on the ratio of pulse tail to total integrals [4]. This PSD parameter is widely used in gauging PSD performance in organic scintillators [5, 6, 7]. There are other methods for quantifying PSD parameters [8], all of which provide a way of clustering neutrons and photons in a particular two dimensional space. Typically one dimension of this space is the PSD parameter, and the other is some metric of energy deposition such as pulse height or pulse integral. A well chosen PSD parameter ensures adequate separation between photon and neutron populations. The width of the photon and neutron distributions is inversely proportional to deposited energy, which makes classification difficult at lower energies. In this paper we apply a low threshold of 25 keVee ("kilo-electron Volt electron equivalent"), equivalent to estimated 245 keV neutron deposited energy.

1.2. New classification method

In this work we present a new classification methodology which departs from typical approaches of drawing the optimal decision boundary between photon

and neutron distributions [9, 10]. In our methodology, instead of segregating pulses into distinct groups, each pulse is assigned a neutron and photon weight based on posterior probability calculated from Bayes' theorem using the photon-to-neutron ratio as the prior probability as will be discussed in Section 3.2. The posterior probabilities are then used to estimate a new prior, and the process repeats until a convergence criteria is met. This iterative scheme successfully adapts the probability space to different proportions of neutrons and photons in the measured data. Furthermore, the sum of posterior probabilities is shown to be a better estimator of total instances of photons and neutrons than an optimal decision boundary that minimizes misclassifications. For our two class problem, the decision boundary that minimizes misclassification is set at a line where posterior probability equals 50%. Events with a PSD parameter above this boundary are classified as neutrons and below as photons.

Performance of the PSD method in this paper was quantified from a neutron detection point of view. Any effective PSD method has to simultaneously maintain high neutron efficiency and a high photon rejection rate [11]. Photon data sets were taken from measurements of pure photon sources, and neutrons from a time correlated measurement of an Am-Be source. Photon and neutron pulse data were combined to form synthetic data sets with varying photon-to-neutron ratios. Neutron efficiency was calculated as a function of this ratio by summing posterior probabilities, we define this as the probability weighted method. This was then compared to neutron efficiency obtained by discrimination using a decision boundary.

2. Experiment

Multiple measurements were performed with the same experimental setup to benchmark the new Bayesian PSD method. An Am-Be source was measured to provide a data set of time tagged neutrons. Three photon sources, ^{137}Cs , ^{60}Co and ^{232}Th were measured separately to provide the corresponding set of pure photon pulses. The photon and neutron data sets were combined to produce

57 mixed data sets with desired photon-to-neutron ratios.

58 This approach was favored over combined measurement of neutron and pho-
59 ton radiation sources for two reasons. First, direct measurement of high photon-
60 to-neutron ratio would necessitate high count rates, which introduces compli-
61 cations from incomplete double pulse rejection. Double pulses are readily char-
62 acterized as neutron pulses, and their inclusion perturbs the metrics used to
63 quantify PSD performance. Secondly, synthetically adding in photon pulses into
64 the mixed data sets gives us greater control in the choice of a photon-to-neutron
65 ratio. Finally, uncertainties associated with the detector-source geometry and
66 source strength, which would be included if mixed radiation fields were mea-
67 sured directly, are precluded from the estimation of photon-to-neutron ratio
68 when data sets are mixed after measurements.

69 *2.1. Set-up and detector settings*

70 Two 2×2 " EJ-309 organic liquid scintillators coupled to Hamamatsu H1949-
71 50 photomultiplier tubes (PMT) were used for the measurements. Full wave-
72 forms were digitized with a CAEN DT5720, 12-bit, 250 MHz digitizer which were
73 saved on a computer through a USB cable. For the Am-Be measurement, one
74 detector was positioned approximately 49.5 cm from the source and was used to
75 benchmark the PSD method; a second detector was positioned approximately
76 0.5 cm from the source and was used to time tag correlated fast neutrons and
77 high energy gammas originating in the (α, n) reaction [12]. The biases applied
78 on the first and second PMTs were -2050 V and -1800 V, respectively. Despite
79 the disparity in the applied voltage, the effective gains were similar because of
80 the use of a 20 dB and 14 dB attenuators with the first and second detectors,
81 respectively.

82 The two detectors were configured in a face-to-face orientation 50 cm apart
83 on an aluminum frame 100 cm from the floor. The 73.7 mCi Am-Be source was
84 placed 0.5 cm away from the second detector for a 108 hour long measurement.
85 The three photon sources measured were 45.7 μCi ^{137}Cs , 54.5 μCi ^{60}Co and 13.5
86 μCi ^{232}Th . These photon sources were measured 20, 25, and 15 cm away from

87 the first detector, respectively. The photon sources were measured at different
 88 distances to keep the count rate constant around 2000 Hz. A picture of the
 89 setup with the Am-Be source in position is shown in Figure 1.

Figure 1: Setup of the two detectors with Am-Be source in position.

90 The position of the Compton edge was estimated at 80% of the peak height,
 91 through linear interpolation between edge peak and baseline. Pulse waveforms
 92 were integrated with a 240 ns integration window. The effective measurement
 93 threshold was 18 keVee, but 25 keVee threshold was used in post processing.
 94 This threshold corresponds to neutron energy deposited on hydrogen of 245 keV,
 95 as calculated from EJ-309 integral light output coefficients [13].

96 3. Method

97 Applying Bayes' theorem requires a conditional probability, or likelihood,
 98 and a prior probability. In this work, we define the former as the value of an
 99 energy dependent Gaussian fit for a given tail-to-total ratio and the latter as the
 100 energy dependent photon-to-neutron ratio. The discussion on estimating both
 101 parameters proceeds in the following two sections. Initially, the prior probability
 102 is not known and thus it must be inferred from the data by an iterative proce-

103 dure. Therefore, the Bayesian probability is adaptive to a particular collection
 104 of data.

105 *3.1. Fitting detector specific parameters*

106 The detector specific parameters were determined from an energy-dependent
 107 double Gaussian fit to the tail to total ratio vs. total integral histogram. This
 108 step would preferably be accomplished on a data-set with nearly equal pho-
 109 ton and neutron populations. The fit provides energy dependant means and
 110 standard deviations for both photons and neutrons. These parameters are only
 111 system dependent, and like gain calibration procedures only need to be per-
 112 formed once for a particular experiment.

113 An open source unconstrained non-linear optimization algorithm was used
 114 for fitting the Gaussian parameters [14]. Each group was fit in descending
 115 order of light output, with the previously calculated coefficients used as the
 116 next initial guess. This ensured continuity of the coefficients across all groups,
 117 and facilitated the precarious fitting at lower light outputs where distributions
 118 overlap the most. The Gaussian distributions were normalized and took on the
 119 familiar form

$$f(s) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(s-\mu)^2}{2\sigma^2}\right) \quad (1)$$

120 where s is the tail-to-total ratio.

121 In order to calculate the Bayesian probability for each pulse individually, the
 122 Gaussian coefficients were fit across light output range of interest. Smoothing
 123 splines were used to approximate the mean and standard deviation coefficients
 124 as a function of light output. The resulting R-squared values were greater than
 125 0.99 for all fitted parameters. The result of the fitting procedure is shown in
 126 Figure 2, as applied to a subset of the Am-Be data. Conditional probabilities
 127 used for PSD analysis in this paper were taken from these fits.

128 By fixing the mean and standard deviation parameters from a calibration
 129 data set we assume they only depend on the detector system, and therefore

Figure 2: The mean (solid lines) and three standard deviation (dashed lines) fits as applied to the Am-Be data set.

do not need to be refitted for other measurements. A comparison of mean and standard deviation fits for the photon sources measured, ^{137}Cs , ^{60}Co and ^{232}Th , has shown this to be the case. In all three cases the standard deviations were nearly identical across the entire light output range. The mean varied near regions of high count rate (i.e., Compton edge), although at most only by 10% of the standard deviation. As a result it is possible to use a single calibration data set — preferably one with nearly equal number of photons and neutrons — to produce the necessary fits.

3.2. Inferring the photon-to-neutron ratio

The Gaussian fits provide the conditional probabilities, but we still require a prior in order to calculate a photon or neutron posterior probability. In our case this is the energy dependant ratio of photon-to-neutrons, which will change depending on the incident radiation (i.e. the type of radiation source measured). Therefore we introduce an iterative procedure that updates the photon-to-neutron ratio by recalculating posterior probabilities until a convergence criteria in the photon and neutron populations is met.

146 The following formulation of the Bayes' theorem was used for calculating
 147 posterior probabilities for either photons

$$P(\gamma|s) = \frac{f_\gamma(s)R_{\gamma/n}}{f_\gamma(s)R_{\gamma/n} + f_n(s)} \quad (2)$$

or neutrons

$$P(n|s) = \frac{f_n(s)}{f_\gamma(s)R_{\gamma/n} + f_n(s)} \quad (3)$$

where $f(s)$ are the Gaussian fits from Eq. 1, the γ or n index indicates the photon and neutron distribution and s is the PSD parameter, in this case the tail-to-total ratio. $R_{\gamma/n}$ is the ratio of the estimated number of counts in the photon and neutron distributions within a light output group. The number of instances of photons and neutrons is estimated by summing the posterior probabilities of each class

$$N_\gamma = \sum_{s \in E_i} P(\gamma|s) \quad (4)$$

$$N_n = \sum_{s \in E_i} P(n|s) \quad (5)$$

148 for a particular light output group E_i .

For the first iteration $R_{\gamma/n}$ is assumed to be unity, then the results from Eqs. 2 and 3 are used to estimate its value for the subsequent iteration:

$$R_{\gamma/n} = \frac{N_\gamma}{N_n}. \quad (6)$$

149 The iterative approach used is an example of an expectation-maximization
 150 algorithm for Gaussian mixtures [15] with fixed means and standard deviations.
 151 Iterations terminate when the convergence criteria is satisfied. Convergence
 152 criteria was defined as 1% difference in total $R_{\gamma/n}$ between two consecutive
 153 iterations. This iterative scheme is robust, and converges to the same solution
 154 given a wide range of initial $R_{\gamma/n}$.

155 3.3. Variance estimation

156 Since the sum of probabilities, not counts, is used to estimate the size of
 157 photon and neutron populations, we added a term in the variance calculation

158 to account for the uncertainty of the posterior probability itself. In this work
 159 the variance on the total number of estimated instances N was calculated by

$$Var(N) = \sum P_i + \sum P_i(1 - P_i) \quad (7)$$

160 where P_i are either photon or neutron posterior probabilities for each pulse. N
 161 represents the number of photon or neutron instances as calculated by Eqs. 4
 162 and 5. The first term in Eq. 7 sums to N and represents the variance on the
 163 total number of counts. The second term is the variance on each individual
 164 probability, which follows a binomial distribution because there are only two
 165 classes, photons and neutrons. In the limit that all the P_i are large the variance
 166 estimation reduces to N . On the other hand, if P_i are small than the variance
 167 estimation is effectively doubled to $2N$.

168 4. Results

169 4.1. Time-tagged neutrons

170 A set of “pure neutron” pulse data was needed to benchmark the Bayesian
 171 PSD method, and it was obtained from time correlated measurement of the
 172 Am-Be source. Figure 3 shows the resulting correlated counts for all particle
 173 pair combinations weighed by correlation probabilities. Eqs. 2 and 3 were used
 174 to calculate the Bayesian probabilities for each detector.

175 The presence of photon-photon correlations at times greater than 12 ns is
 176 due to the production of photons induced by high energy neutrons. Only one
 177 correlated neutron is produced from the (α, n) reaction, however the presence
 178 of neutron-neutron correlations indicates significant contributions from both
 179 scatter and $(n, 2n)$ reactions in Beryllium.

180 The neutron data set was taken from the pulses with time correlations be-
 181 tween 15 and 50 ns, which corresponds to neutron energies of 5.6 ± 0.4 MeV
 182 and 460 ± 19 keV given detector separation of 50 cm. We approximated 96.5%
 183 neutron purity, for a total of 105,000 total neutron counts, in the time-tagged
 184 neutron data set by applying the PSD approach presented in this paper.

Figure 3: Timing distribution of correlated counts between two liquid scintillator detector using and Am-Be source. The minimum correlation probability threshold was 99% to minimize appearance of misclassified correlations.

185 4.2. Neutron acceptance

186 The neutron acceptance rate is defined relative to the counts found within the
 187 timing window in the Am-Be measurement before mixing in photons from the
 188 gamma source data sets. Photon pulses from ^{137}Cs , ^{60}Co and ^{232}Th measure-
 189 ments were mixed into the neutron data with estimated total photon-to-neutron
 190 proportions ranging from 1 to 2000. An example of the neutron probability map
 191 from a 1:1 mixture with ^{60}Co data is shown in Figure 4. Our iterative approach
 192 ensures that the neutron probability decreases as more photons are added to
 193 the mixed set.

194 Figures 5 and 6 show the photon and neutron distribution before being mixed
 195 and after separation for 1:1 and 1000:1 photon-to-neutron ratios, respectively.
 196 In the 1:1 mixture the neutron distribution shape and magnitude is persevered
 197 after mixing, however the number of photons has increased because of the pho-
 198 tons present in the time-tagged neutron data set. In the 1000:1 mixture the
 199 contribution of those photons is relatively much smaller, and as a result the
 200 photon distributions before and after mixing appear nearly identical. Due to

Figure 4: The distribution of neutron posterior probabilities for 1:1 mixture of ^{60}Co and time tagged neutron data (color available online).

the higher number of photons present in the 1000:1 mixture, the neutron probabilities were reduced causing a drop in efficiency, particularly at low energies. The drop in neutron acceptance rate at low energies is demonstrated in Figure 7, where neutron acceptance rate is presented for each light output group. Only 10% of the available neutrons, approximately 10,500 out of the set of 105,000, were used in the 1000:1 mixture because of the limited number (~ 15 million) of photon events available from the gamma source measurements.

The neutron acceptance rate as a function of photon-to-neutron ratio is shown in Figure 8. The neutron acceptance rate shown is integrated over all light output groups, essentially the integrated result shown in Figure 7 but for all gamma sources. The neutron counts were weighted by their neutron posterior probabilities, from Eq. 3, which we call the probability weights technique. Since neutron posterior probabilities are calculated for every event we can sum them to estimate the total population of neutrons. This was compared to the integral of counts above the decision boundary defined in Section 1.2, where every event is counted as either a neutron or a photon. For both methods the same iterative procedure, described in Section 3.2, was used to approximate the

Figure 5: The distribution of time-tagged neutrons and ^{232}Th photons before and after mixing in a 1:1 ratio.

Figure 6: The distribution of time-tagged neutrons and ^{232}Th photons before and after mixing in a 1000:1 ratio.

Figure 7: Neutron acceptance rate for the ^{232}Th data set as a function of light output for different photon-to-neutron ratios.

218 prior distribution $R_{\gamma/n}$. As expected, the summation of posterior probabilities
 219 maintains an acceptance rate up to 9% higher than those obtained using decision
 220 boundary discrimination.

221 Regardless of the photon-to-neutron ratio, the integrated neutron acceptance
 222 rate was kept below 100% because of the iterative approach that adjusted the
 223 photon-to-neutron ratio for each data set. At some light output values the
 224 acceptance rate did exceed 100% but only within a few percent of the marginal
 225 error. Traditionally the decision boundary is set based on a calibration data set
 226 and is then used for subsequent measurements [9]. This traditional approach is
 227 compared to our iterative technique in Figure 9. The static decision boundary
 228 was chosen assuming a photon-to-neutron ratio of 1. The problem with using a
 229 static boundary is that photons always have some probability of being above it
 230 in the neutron region. The neutron acceptance rate will increase linearly with
 231 the photon-to-neutron ratio as more photons are added into the mix.

Figure 8: Neutron acceptance rate as a function of photon-to-neutron ratio for all three photon sources. Only 10% of the available neutron had to be used to achieve ratios greater than 100, which results in larger error estimations.

Figure 9: Neutron acceptance rate for the ^{137}Cs data set with the static decision boundary chosen assuming photon-to-neutron ratio of 1.

5. Conclusions

The applicability of Bayes' theorem to pulse shape discrimination was demonstrated with the use of a charge integration technique. An iterative approach that infers the ratio of photon-to-neutron light output distributions from data is demonstrated as a robust method for adjusting the probability space for measurements with different photon-to-neutron ratios. Neutron acceptance rate of 90% was maintained with photon-to-neutron ratio of up to 2000. Overall the neutron acceptance rate was up to 9% higher with the probability weighted technique compared with drawing a decision boundary.

The Bayesian PSD technique can be used to calculate neutron and photon probabilities on an event by event basis. For correlation experiments with multiple detectors, correlation probabilities for each possible event type combination can be calculated. These correlation probabilities can be used to set new thresholds to clean-up misclassification rates to a desired level without raising the energy threshold on all events.

247 **Acknowledgments**

248 The author would like to thank Kyle Polack, Marek Flaska, and Aaron Bevill
249 for their feedback regarding the work presented in this paper.

250 This material is based upon work supported by the U.S. Department of
251 Homeland Security under Grant Award Number, 2012-DN-130-NF0001. The
252 views and conclusions contained in this document are those of the authors and
253 should not be interpreted as representing the official policies, either expressed
254 or implied, of the U.S. Department of Homeland Security.

255 Sandia National Laboratories is a multi-program laboratory managed and
256 operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin
257 Corporation, for the U.S. Department of Energys National Nuclear Security
258 Administration under Contract DE-AC04-94AL85000. SAND Number 2015-
259 1190 J.

260 **References**

- 261 [1] G. F. Knoll, Radiation Detection and Measurement, 3rd Edition, John
262 Wiley & Sons, Inc., 2000.
- 263 [2] N. Kornilov, V. Khriatchkov, M. Dunaev, A. Kagalenko, N. Semenova,
264 V. Demenkov, A. Plompen, Neutron spectroscopy with fast waveform
265 digitizer, Nuclear Instruments and Methods in Physics Research Sec-
266 tion A 497 (23) (2003) 467 – 478. doi:[http://dx.doi.org/10.1016/](http://dx.doi.org/10.1016/S0168-9002(02)01790-4)
267 [S0168-9002\(02\)01790-4](http://dx.doi.org/10.1016/S0168-9002(02)01790-4).
- 268 [3] D. A. Shea, D. Morgan, The helium-3 shortage: Supply, demand, and
269 options for congress, Tech. Rep. R41419, Congressional Research Service
270 (2010).
- 271 [4] J. Adams, G. White, A versatile pulse shape discriminator for charged
272 particle separation and its application to fast neutron time-of-flight spec-
273 troscopy, Nuclear Instruments and Methods 156 (3) (1978) 459 – 476.
274 doi:[http://dx.doi.org/10.1016/0029-554X\(78\)90746-2](http://dx.doi.org/10.1016/0029-554X(78)90746-2).

- [5] I. A. Pawelczak, S. A. Ouedraogo, A. M. Glenn, R. E. Wurtz, L. F. Nakae, Studies of neutron γ pulse shape discrimination in ej-309 liquid scintillator using charge integration method, Nuclear Instruments and Methods in Physics Research Section A 711 (2013) 21 – 26. doi:<http://dx.doi.org/10.1016/j.nima.2013.01.028>.
- [6] C. Liao, H. Yang, n/ γ pulse shape discrimination comparison of ej301 and ej339a liquid scintillation detectors, Annals of Nuclear Energy 69 (2014) 57 – 61. doi:<http://dx.doi.org/10.1016/j.anucene.2014.01.039>.
- [7] S. A. Pozzi, M. M. Bourne, S. D. Clarke, Pulse shape discrimination in the plastic scintillator ej-299-33, Nuclear Instruments and Methods in Physics Research Section A 723 (2013) 19 – 23. doi:<http://dx.doi.org/10.1016/j.nima.2013.04.085>.
- [8] K. A. A. Gamage, M. J. Joyce, N. P. Hawkes, A comparison of four different digital algorithms for pulse-shape discrimination in fast scintillators, Nuclear Instruments and Methods in Physics Research Section A 642 (1) (2011) 78 – 83. doi:<http://dx.doi.org/10.1016/j.nima.2011.03.065>.
- [9] A. C. Kaplan, M. Flaska, A. Enqvist, J. L. Dolan, S. A. Pozzi, Ej-309 pulse shape discrimination performance with a high gamma-ray-to-neutron ratio and low threshold, Nuclear Instruments and Methods in Physics Research Section A 729 (2013) 463 – 468. doi:<http://dx.doi.org/10.1016/j.nima.2013.07.081>.
- [10] J. K. Polack, M. Flaska, A. Enqvist, S. A. Pozzi, A computer-aided, visual, charge-integration, pulse-shape-discrimination method for organic scintillators, in: Proceedings of the Institute of Nuclear Materials Management 54th Annual Meeting, Palm Desert, California, 2013.
- [11] R. T. Kauzes, J. R. Ely, A. T. Lintereur, D. L. Stephens, Neutron detector insensitivity criteria, Tech. Rep. PNNL-18903, Pacific Northwest National Laboratory (2009).

- [12] H. R. Vega-Carrillo, E. Manzanares-Acua, A. M. Becerra-Ferreiro, A. Carrillo-Nuez, Neutron and gamma-ray spectra of ^{239}Pu and ^{241}Am , *Applied Radiation and Isotopes* 57 (2) (2002) 167 – 170. doi:
[http://dx.doi.org/10.1016/S0969-8043\(02\)00083-0](http://dx.doi.org/10.1016/S0969-8043(02)00083-0).
- [13] C. C. Lawrence, M. Febbraro, T. N. Massey, M. Flaska, F. Becchetti, S. A. Pozzi, Neutron response characterization for an ej299-33 plastic scintillation detector, *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 759 (0) (2014) 16 – 22. doi:<http://dx.doi.org/10.1016/j.nima.2014.04.062>.
- [14] T. O’Haver, Peak fitter version 5.4 (2014).
URL <http://terpconnect.umd.edu/~toh/spectrum/InteractivePeakFitter.htm>
- [15] Y. Uchida, E. Takada, A. Fujisaki, M. Isobe, K. Ogawa, K. Shinohara, H. Tomita, J. Kawarabayashi, T. Iguchi, A study on fast digital discrimination of neutron and gamma-ray for improvement neutron emission profile measurement, *The Review of Scientific Instruments* 85 (11) (2014) 11E118. doi:10.1063/1.4891711.