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Implementation of Advanced Matrix Corrections for Active Interrogation of Waste Drums Using the CTEN Instrument

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ABSTRACT

The combined thermal/epithermal neutron instrument (CTEN) was designed at Los Alamos to improve measurement accuracy and mitigate self shielding effects inherent in the differential dieaway technique (DDT). A major goal in this research effort has been the development of a calibration technique that incorporates recently developed matrix and self-shielding corrections using data generated from additional detectors and new acquisition techniques. A comprehensive data set containing both active and passive measurements was generated using 26 different matrices and comprising a total of 1400 measurements. In all, 31 flux-and matrix-dependent parameters, 24 positional parameters, two dieaway times, and a correlated ratio were determined from each of the over 1400 measurements. A reduced list of matrix indicators, prioritized using the alternating conditional expectation (ACE) algorithm, was used to train a neural network using a generalized regression technique (GRNN) to determine matrix- and position-corrected calibration factors. This paper describes the experimental, analytical, and empirical techniques used to determine the corrected calibration factor for an unknown waste drum. Results from a range of cases are compared with those obtained using a mobile DDT instrument and traditional DDT algorithms.

INTRODUCTION

Adaptation of the dieaway technique¹ to an assay instrument capable of measuring the fissile content of 55 gallon drums began at Los Alamos National Laboratory in the 1970s. The technique uses thermalized neutrons from a 14-MeV pulsed source to irradiate radioactive waste drums. Prompt neutrons from induced fissions are detected and thus provide a direct measure of the fissile content of the drum. Depending on the contents of the drum, measurement times are from 40 to 200 seconds, with ²³⁹Pu sensitivities ranging from one to 50 milligrams. Although the technique has proved extremely sensitive to fissile material within waste containers, quantitative measurements require corrections to compensate for the effects of the matrix, spatial distribution, and the physical form of the fissile material. Absorption and moderation by the matrix change both the intensity and the slowing-down profile of the interrogating flux. The induced prompt fission neutrons are also moderated, and to a lesser degree absorbed, by the matrix materials. Thus, the fraction of signal neutrons that reach the detectors changes with matrix type, resulting in variations in the overall detection efficiency. To complicate the problem further, matrix effects on both the interrogating and induced-fission signals also depend upon location within the drum. In hydrogencontaining drums, these two effects are directly opposed and at some hydrogen density will tend to cancel

each other, i.e., a position with an increase in interrogating flux, and therefore an increase in the number of fission reactions, will also exhibit a corresponding decrease in signal transmission.

The CTEN design differs from traditional DDT units by using graphite instead of polyethylene in the assay chamber walls. Since the neutron slowing down time in graphite is an order of magnitude greater than in hydrogen, the system thermalizes neutrons at a slower rate. To take advantage of the information available at earlier times after the burst, all signals are provided simultaneously to two independent data collection paths known as the scaler-mode and list-mode acquisitions. During an active interrogation, the scaler mode uses time-gated scalers to collect data from detector signals in five separate windows. The active measurement uses 39 separate ³He proportional counters in 13 shielded detector packages to measure the fissile signal. In addition to the fissile signal, signals from a variety of bare and shielded flux assay monitors, the cadmium-collimated drum monitor, ⁴He detectors and an external flux monitor are also recorded during each of five time-gated windows. During an assay, the drum rotates 360 degrees which is divided into a specified number of angle segments. To enhance the spatial information, data is collected by drum angle as well as by detector package(s). List-mode acquisition is accomplished by routing signals into one of two pulse arrival time recording modules (PATRM)2. Neutron arrival times are recorded with increasing time values with the reference time zero being the start of the burst. Timing data is recorded independently for the sum of the shielded detectors and the cadmium-collimated drum monitor. The drum monitors are collimated such that only neutrons originating from the drum are detected. Thus the dieaway time of the originating neutron flux can be calculated indirectly using the net fissile signal and more directly using the response of the drum flux monitor. The list mode data from the shielded detectors is also used to extract correlations in the neutron signals using the Feynman reduced-variance method³.

DEVELOPMENT OF MATRIX CORRECTIONS

Neutron induced reactions take place at a rate that is directly proportional to both the total number of target nuclei and the scalar neutron flux. A straightforward application of this principle to an empty drum provides the basic calibration factor, K_A (g ²³⁹Pu/net signal). Since both the intensity of the interrogating flux and the transmission of the induced fission neutrons varies with matrix type and source position, corrections to this simple proportionality must be made before an accurate value can be calculated.

The active calibration process described in this paper began with the generation of a comprehensive data set containing both active and passive measurements using 26 different matrices. These standard matrices spanned the range of moderator and absorber densities expected in waste drums by using combinations of hydrogen and boron. Over 1400 measurements were made generating scalar and list mode data as a function of 20 positions within the drum. Data was also collected as a function of drum rotation angle. In addition to the raw signals, i.e., net counts from 32 independent detector channels in five

time windows and timing data for the shielded detectors and drum flux monitor, several related quantities were calculated. These included ratios of detector signals from various positions and from different time windows, Feynman distributions, correlated ratios, dieaway times, and detector variances. In all, 26 flux-and transmission-dependent parameters, 22 positional parameters, 2 dieaway times, and a correlated ratio were determined for each of the over 1400 measurements. A calibration factor was calculated for each position thus providing the required correction factor, relative to an empty drum, for that matrix and position. The measured transmission was used to separate the transmission- and flux-related correction factors. The next task for this process was to determine which quantities were most indicative of the correction factor. To expedite this prioritization, the ACE algorithm was used.

The ACE algorithm⁴ is suited for experimental situations where the system response has a complicated dependence on many measurable parameters, i.e., a multivariate problem. The algorithm generates a functional form and performs a numerical regression using only empirical data. As various combinations of parameters were evaluated, the span of the ACE-calculated transformations was a good indicator of what variables contributed most to the known correction factor. We used this technique to determine three main indicators: 1) interrogating flux generation and decay, 2) fissile signal transmission, and 3) position of source material within the matrix. In the final analysis, the same two parameters were chosen to determine flux and transmission correction factors, i.e., the ratio of shielded flux monitors from the epithermal time regions to the bare flux monitor from the thermal region (SFM/BFM), and the normalized drum monitor response from the thermal region (DFM/EFM). While other indicators were in some cases stronger, i.e., the correlated ratio for transmission, these were chosen to provide a correction factor for all matrices independent of the amount of source material present. Since these indicators are a function of the interrogating source, the statistics are always good. For determining source material position, the best indication of source height within the drum was the ratio of the shielded detectors located in the ceiling to those in the floor (TOP2BOT). The best radial indicator is the ratio of the sum of the squared deviations from the average of the individual angle responses to the squared average response for detectors located in the assay walls (SUMDEV).

In the development of the original DDT active algorithms, the total correction factor was divided into moderator and absorption correction factors. Thus the moderator correction factor included the effects of the moderator on both the interrogating flux and the transmitted fission neutrons, which are directly opposite. This measurement paradigm further complicates the relationship between measured quantities and the neutronic properties of the matrix. We chose instead to determine the overall matrix effects on the interrogating thermal flux independently of the effects of the transmitted fission neutrons. To accomplish this, we used a generalized regression neural network (GRNN)⁵ to first determine position-averaged correction factors. For the average flux correction factor, position averaged values for SFM/BFM, DFM/EFM, and the known flux correction factor were used to train the a neural network. For the average transmission correction factor, the same indicators were used to train a neural network with the known

transmission correction factor as the desired output. Thus, the number of cases for these neural networks was equal to the number of drums (26). These position-averaged correction factors along with the two position indicators, TOP2BOT AND SUMDEV, were then used as input to train neural networks with the desired outputs as the known position-sensitive flux and transmission correction factors. Thus, for each correction factor, two neural networks were independently trained. Since all data points were used to train for position-sensitive correction factors, 520 cases were used in all. Figures 1 and 2 compare the known flux and transmission correction factors to those determined by the GRNN method. The total correction factor is the product of the flux and transmission correction factors. Figure 3 compares the known and predicted overall correction factors.

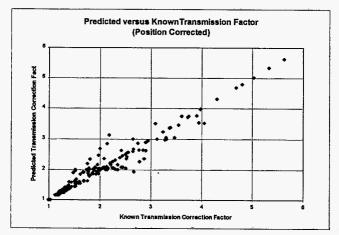


Figure 1. Comparison of the known transmission correction factor with the transmission correction factor determined via the GRNN method.

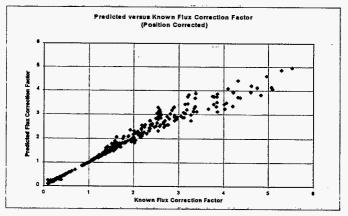


Figure 2. Comparison of the known flux correction factor with the flux correction factor as determined via the GRNN method.

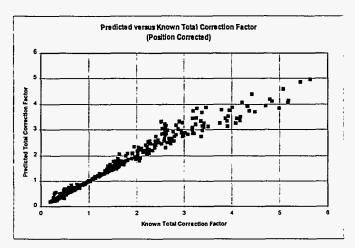


Figure 3. Comparison of the known total correction factor with the prediction. The predicted total correction factor is the product of the predicted transmission correction factor and predicted flux correction factors.

| Matrix Properties | | Uncorrected Mass | | Corrected Mass | |
|-------------------|---------------|------------------|---------|----------------|---------|
| Hydrogen | Boron Density | Average Mass | Std Dev | Average Mass | Std Dev |
| Density (g/cm³) | (g/cm³) | (mg) | (%) | (mg) | (%) |
| 0.00536 | - | 668 | 18.7 | 359 | 2.6 |
| 0.0122 | - | 957 | 20.2 | 348 | 8.5 |
| 0.0133 | - | 1015 | 24.1 | 354 | 10.4 |
| 0.0193 | - | 1253 | 25.5 | 361 | 5.5 |
| 0.0222 | • | 1305 | 25.9 | 389 | 6.7 |
| 0.0342 | - | 841 | 21.0 | 377 | 12.7 |
| 0.0381 | - | 691 | 18.5 | 378 | 15.5 |
| 0.0414 | - | 509 | 13.4 | 364 | 4.5 |
| 0.0450 | - | 495 | 14.3 | 365 | 5.2 |
| 0.00942 | - | 392 | 6.1 | 359 | 1.0 |
| 0.00114 | 2.532E-5 | 313 | 6.4 | 361 | 1.3 |
| 0.001153 | 5.5064E-5 | 282 | 5.5 | 358 | 1.8 |
| 0.001179 | 1.1013E-4 | 240 | 4.6 | 355 | 3.6 |
| 0.001667 | 1.6519E-4 | 263 | 14.9 | 364 | 2.9 |
| 0.001879 | 2.2025E-4 | 212 | 17.4 | 370 | 6.6 |
| 0.002297 | 2.4779E-4 | 198 | 20.5 | 370 | 6.5 |
| 0.002441 | 3.0285E-4 | 187 | 26.9 | 367 | 9.1 |
| 0.002443 | 3.5798E-4 | 161 | 25.5 | 372 | 11.6 |
| 0.002419 | 4.1298E-4 | 135 | 25.7 | 367 | 9.9 |
| 0.002444 | 4.6804E-4 | 121 | 25.9 | 348 | 10.2 |
| 0.002502 | 5.5064E-4 | 106 | 28.4 | 332 | 11.8 |
| Iron Scrap | - | 135 | 14.1 | 386 | 11.0 |
| | Average | 476 mg | 18.4% | 364 mg | 7.2% |
| | Std Dev (%) | 79.7 % | | 3.5 % | |

Table 1. In this set of measurements, 361 mg of ²³⁹Pu was measured in 20 different positions for each matrix. The average mass and standard deviation for each of the 20 positions for both uncorrected and corrected assays is shown. The bottom row is the relative standard deviation for the average uncorrected and corrected masses of the different matrices.

RESULTS

Table 1 presents both uncorrected and corrected results for both moderating and absorbing matrices. The hydrogen density varies from 0 to 0.045 g/cm³, roughly equivalent to 40% water by weight. In this set of measurements, 361 mg of ²³⁹Pu was measured at 20 different positions within each matrix. The average measured value for each matrix and the standard deviation for the 20 positions is given. The overall average positional standard deviation drops from 18.4% to an average of 7.2% for the corrected mass values. Without a correction, the relative standard deviation in the average calculated mass from the set of matrices is 80% dropping to less than 4% using both position-sensitive transmission and flux correction factors. Table 2 compares comparable matrices measured with the CTEN instrument using the GRNN method and the mobile DDT instrument using traditional algorithms.⁶ Since the traditional DDT algorithms assume a homogenous matrix with a distributed source, they are unable to correct for matrices containing moderately high levels of moderator or absorber. While the variation with position improves in the traditional algorithms for benign matrices, the measured value is underestimated, i.e., measured/true mass equal to 83% with a 2.2% positional standard deviation.

| Matrix Properties | | Mobile DDT-Corrected | | CTEN-Corrected | |
|-----------------------------|--------------------------|----------------------------------|----------------|----------------------------------|----------------|
| Hydrogen Density (g/cm³) | Boron Density (g/cm³) | Average Measured/True Mass | Std Dev (%) | Average Measured/True Mass | Std Dev (%) |
| 0.00536 | - | 0.92 | 11.8 | 1.00 | 2.6 |
| 0.0342 | - | 1.03 | 44.9 | 1.03 | 14.4 |
| 0.00942 | - | 0.93 | 1.6 | 0.99 | 1.0 |
| 0.001667 | 1.6519E-4 | 0.83 | 2.2 | 1.00 | 2.8 |
| 0.002441 | 3.0285E-4 | 0.88 | 14.4 | 1.02 | 9.5 |
| 0.002502 | 5.5064E-4 | 0.90 | 21.3 | 0.93 | 11.0 |
| Iron Scrap | - | 1.05 | 13.1 | 1.07 | 11.0 |
| | Average | 0.93 | 15.7 % | 1.00 | 6.7% |

Table 2. Comparison of results from the mobile DDT and CTEN instruments for similar matrices. Since the center radial point was not measured with the mobile DDT instrument, the standard deviation is over 15 data points.

SUMMARY

We have used a generalized regression network to develop independent corrections for interrogating flux and fissile neutron transmission variations as a function of position. These corrections have resulted in reducing positional variations over the traditional algorithms used in DDT. While a standard set of matrices were used to train the neural networks, other matrices can easily be incorporated into the trained network. Work in the future will concentrate on expanding the range of validity of the neural network through measurements of both mock and real waste drums.

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