ANS RPSD 2014 - 18th Topical Meeting of the Radiation Protection & Shielding Division of ANS
Knoxville, TN, September 14 – 18, 2014, on CD-ROM, American Nuclear Society, LaGrange Park, IL (2014)

Identification of Leakage Spectra from Neutron Activation in Glass/Cherenkov Detectors

Douglas E. Peplow, Zane W. Bell and Steven P. Hamilton

Oak Ridge National Laboratory, P.O. Box 2008, MS-6170, Oak Ridge, TN, 37831, USA, peplowde@ornl.gov

INTRODUCTION

Oak Ridge National Laboratory has developed a new detector system concept that can be deployed in major US cities to identify the leakage spectrum of a nuclear weapon detonation. The detector is based on a small piece of glass that has been doped with an element that has a high activation cross section and creates a radioactive activation product with a reasonable length half-life. The decay of the activation product(s) after exposure creates Cherenkov light which can propagate to a photomultiplier and can be turned into a count-rate signal. The signal from each glass in each detector in a city can be individually analyzed to determine the amount of activation products produced in the initial exposure. Several activation products can be determined in a single glass if the half-lives are not the same.

With a known source location and a detailed 3D model of a city, neutron transport simulations can be made by using a library of known sources to predict the activation amounts each source would have made in the glass detectors. A simple comparison of the measured activation amounts and the predicted activation amounts can identify the source spectrum contained in the library that is the best match. Choosing dopants with a large range in the threshold energy for the activation reactions can help to differentiate the spectra in the library.

The method of matching the measured activation amounts to the predicted activation amounts to identify the source spectrum is similar to the analysis used for the passive Activation Foil Integrated Detector System (AFIDS [1]), also developed at Oak Ridge National Laboratory. In AFIDS, wires or foils of known mass and isotopic composition are deployed before and then collected after exposure. Counting gammas in a nearby laboratory determines the activation amounts. The glass/Cherenkov system is active—it uses a battery-powered detector to count flashes of Cherenkov light and to communicate its count rates to a remote location for further analysis.

Work has been performed in several areas, including glass making, automated detection and communication systems and software tools for analysis of the signal and source spectrum identification. Bell and Boatner [2,3] have developed phosphate glasses that incorporate elements with high capture cross sections, and Hayward [4,5] has simulated the response with Geant4 and measured the response with isotopic gamma sources. This paper will focus on the software tools for determining the activation amounts from the Cherenkov count-rate signal and finding the best matching source spectrum. A test problem in an urban environment is used to demonstrate the potential of a spectrum identification system using neutron activation.

METHODS

Determination of Activation Amounts

The count-rate signal from the detector is the sum of the individual contributions from each decaying activation product. The total count rate $C(t)$, in counts/sec, as a function of time, t, since the instantaneous exposure from the source can be modeled as

$$C(t) = \sum_{j}^{J} \kappa_j \lambda_j x_j e^{-\lambda_j t} \ , \tag{1}$$

with
- J number of activated products
- x_j number of initial atoms of activation product j
- λ_j decay constant of activation product j [/sec]
- κ_j Cherenkov conversion constant (counts per decay of activation product j)

To unfold this signal and determine the number of initial atoms of each activation product (the vector $\vec{x}$), a linear least squares approach can be used. The measured average count rate c_i in time bin i between times t_{i-1} and t_i (with $t_0 = 0$ and $\Delta t_i = t_i - t_{i-1}$) is then

$$c_i = \frac{1}{\Delta t_i} \sum_{j}^{J} \kappa_j x_j \left(1 - e^{-\lambda_j \Delta t_i}\right) e^{-\lambda_j t_{i-1}} \ . \tag{2}$$

For each activation product j, a basis function representing the count rate in each time bin i per initial product isotope atom can be defined to be

[1] Notice: This manuscript has been authored by UT-Battelle, LLC, under contract DE-AC0500OR22725 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

$$f_{ij} = \frac{1}{\Delta t_i} \sum_j^J \kappa_j \left(1 - e^{-\lambda_j \Delta t_i}\right) e^{-\lambda_j t_{i-1}}, \tag{3}$$

such that $c_i = \sum_j^J x_j f_{ij}$. With I time bins ($I \gg J$), the vector of initial isotope amounts, $\vec{x}$, is then the solution to an overdetermined system of linear equations equating the model of the count rate to each measured value of the count rate c_i.

$$\begin{bmatrix} f_{11} & f_{12} & f_{13} & \cdots & f_{1J} \\ f_{21} & f_{22} & f_{23} & \cdots & f_{2J} \\ f_{31} & f_{32} & f_{33} & \cdots & f_{3J} \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ f_{I1} & f_{I2} & f_{I3} & \cdots & f_{IJ} \end{bmatrix} \begin{bmatrix} x_1 \\ x_2 \\ x_3 \\ \vdots \\ x_J \end{bmatrix} = \begin{bmatrix} c_1 \\ c_2 \\ c_3 \\ \vdots \\ c_I \end{bmatrix} \tag{4}$$

In matrix notation, this can be expressed as $F\vec{x} = \vec{c}$. Overdetermined systems like this are usually solved by pre-multiplying both sides of the above equation by the transpose of the matrix F giving $A\vec{x} = \vec{b}$, which is the familiar square ($J \times J$) system of normal equations with $A = F^T F$ and $\vec{b} = F^T \vec{c}$. In this project, the matrix F can have a rather large condition number, which in turn makes matrix A have a very large condition number, subjecting the uncertainty estimate of the solution $\vec{x}$ to large numerical instabilities. For this project, the system $F\vec{x} = \vec{c}$ is solved using a QR factorization routine from LAPACK and the uncertainty estimates in $\vec{x}$ are much more reliable.

Spectrum Matching

From the neutron transport simulations using each of the source spectra in the library, the predicted numbers of activation products, B_{sdj}, for source spectra s, detector location d and activation product j can be computed. Associated with these predicted activation amounts are the uncertainties u_{sdj}, based on the composition uncertainties in the glass material weight fractions. The measured values of the initial activation amounts and their uncertainties, $M_{dj} \pm \sigma_{dj}$, are compared to the predicted amounts in a two-step process to find the yield and the best matching source spectrum.

Note that predictions only need to be made for the initial activation amounts in the detector location and glass tube combinations that were actually measured. In the methods described below, the sums are only over those initial amounts that have been measured ($M_{dj} > 0$).

The first step in the analysis is to determine the yield for each library source spectrum that would be necessary to make that spectrum's predicted activation amounts match the measured data as accurately as possible. For each source spectrum s, the yield, y_s, is found by minimizing the χ^2 quantity of

$$\chi_s^2 = \sum_d^D \sum_j^J \frac{\left(y_s B_{sdj} - M_{dj}\right)^2}{y_s^2 u_{sdj}^2 + \sigma_{dj}^2}. \tag{5}$$

Due to the presence of the yield in the denominator, it is difficult to analytically solve Eq. (5) for the yield. An iterative approach can be used to estimate the yield without uncertainties in the predicted amounts, then using that yield to compute the total uncertainty term, then estimating the yield again. Once a yield has been determined for each library source spectrum, the final χ_s^2 value is computed using Eq. (5). Each set of predicted activation amounts can be compared to measured amounts, and the accuracy of the predictions can be ranked.

One source spectrum will have a χ^2 value that is lower than the others, indicating that it is the best match. The magnitude of a χ^2 value provides some information about how well that source spectrum matches the measured data. A good match should have a χ^2 value less than or equal to the number of degrees of freedom (d.o.f., the number of measured values with $M_{dj} > 0$). The statistical P-value (the probability that the distribution assumes a χ^2 value greater than the measured value χ_s^2) can also be calculated for each library source spectrum. If the best match has a high χ^2 value ($\chi^2 \gg$ d.o.f.), this indicates a poor match—meaning that the true source that caused the measured activations is not in the source library.

Event Analysis

Prior to the event, data in the following areas would be prepared: a detailed geometry model of the city, ready to be used in transport simulations; a list of the locations of the detectors in the city model; the compositions of the glasses in each detector; and a library of different types of source spectra, ready to be used in neutron transport simulations.

After the event, a good estimate of the source location is assumed to be known. Simulations using every source spectrum in the library at that source location are then made to predict the initial activation amounts in each glass at each detector location. These simulations are done once and stored.

Immediately after the event, count-rate signals from the surviving detectors come into the analysis center. The count rates are unfolded to determine the number of activation products produced during the exposure. Soon after the event, it will only be possible to determine activation products with short half-lives. As more time elapses, it will be possible to determine longer-lived

activation product amounts. The process of determining the initial activation amounts and their uncertainties only takes a few seconds.

After the transport simulations are complete, the predicted activation amounts are compared to the measured amounts to determine the best matching source spectrum and its yield. This process also takes only a few seconds. The combination of determining the actual amounts of activation products in the glass and finding the best match in the simulated amounts can be repeated many times as more count-rate information arrives at the analysis center. Source identification and the yield estimates will become more reliable as more actual activation amounts are determined from the count-rate signals at longer times from the event.

DEMONSTRATION

Pre-Event Preparation

A test problem based on a 3 × 3 km section of Manhattan (Fig. 1 [6]) was used to demonstrate the methods for an event analysis. The city model used 5-m^3 voxels of air, low-density concrete (buildings) and soil. To save time and memory for the neutron transport simulations, the resolution was reduced to voxels of 10 × 10 × 5 m. A library of source spectra was created using Fat Man and Little Boy [7], a 14-MeV source, and three fission (Watt) spectra. Several linear combinations of the 14-MeV source and the others were added to make a library of 18 spectra. Ten glasses (such as SiO_2, MgF_2, and lead/phosphorus glasses doped with Na, Mn, Mg, In, or Sc) were used in each detector location. Each element in the glass was assumed to have a 1% uncertainty in its weight fraction. A detector with all ten glasses was placed at the center of each 0.5-km grid square. Some detectors were atop buildings and others were at ground level. Detectors were not used in the two grids in the north-west section of the model that corresponded to the Hudson River. In all, 34 detector locations with 10 glasses each were used. Each glass contained ~10 activation products of different half-lives.

To create 'measured' data, one of the source spectra (Source 2) in the library was chosen as the 'real source,' with a yield of 10 kT. The source location was defined as ground level in Times Square. Initial activation amounts were computed for all of the glasses at all of the detector locations. These initial amounts included an amount of statistical noise based on the number of neutrons arriving and interacting in an activation reaction. Decay of the activation products was simulated, and count rates were recorded in 40-s intervals for two weeks, again including the addition of appropriate statistical noise.

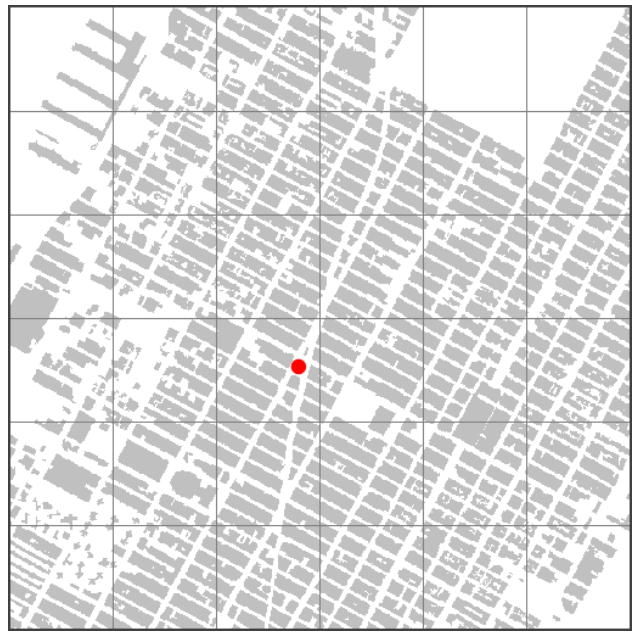

Fig. 1. Model of a 3 × 3 km section of central Manhattan with a detector (with ten glasses each) at the center of every 0.5 km^2. The red circle represents the source location at Times Square.

Post-Event Analysis

For the post-event analysis, each of the 18 source spectra was used in a neutron transport calculation to predict the 3502 activation amounts in all of the glasses at each detector location. The process of determining the actual amounts of activation products and the source spectra and yields that were the best matches was done at three times after the event: 1 day, 3.5 days and 14 days.

At 1 day after the event, 538 activation amounts were determined from the 'measured' count-rate data. Comparing to the 18 sets of predicted amounts, the yield for each was determined and a χ^2 value was computed. The best χ^2 value was 0.95 (or 0.0018 per d.o.f.), which was for Source 2—the correct match. The yield was computed to be 10 kT—the correct yield. The second best match was for Source 17, which has a very similar spectral shape to Source 2, with a χ^2 value of 41 (0.076 per d.o.f). Both of these sources are considered to be good matches. The third best match had a χ^2 value of 6000 (11 per d.o.f), which is a very poor match. The other 15 source spectra also matched poorly, with χ^2 values ranging from 230,000 to 2.9×10^6.

With 3.5 days of count-rate data, 756 initial activation amounts were determined from the count-rate signals. When the matching program was run, Source 2 and Source 17 were good matches again, with χ^2 values of 3.7 and 63. The next best match had a χ^2 value of 8300, indicating a poor match.

After 14 days, 927 initial activation amounts could be determined. The lowest χ^2 values from the matching were 150 (or 0.16 per d.o.f) and 230 (0.25 per d.o.f.), corresponding to Sources 2 and 17. The yield for Source 2 was computed and again matched the correct yield. The next closest match had a χ^2 value of 10000—a very poor match.

Other Variations

Choosing another source spectrum to use for simulating the 'measured data' gave similar results—the best matching spectrum was always the correct source, with similar shaped source distributions scoring as second and third best. Yield estimates matched the yield that was used to produce the measured data. The χ^2 values clearly indicated that the correct source spectrum was a good match and the other spectra were not.

Tests were also performed that assumed that detectors close to the source were destroyed and unable to return count-rate data to the analysis center. The overall results were unchanged—the correct source was selected as the best match, and the correct yield was computed.

SUMMARY

The analysis methods developed to identify the neutron leakage spectra from Cherenkov light given off by neutron activation in doped glasses work well. The calculation used to unfold the count-rate information into the number of actual activation products uses robust algorithms that can also compute associated uncertainties. These uncertainties and the known uncertainties of the glass tube material compositions are incorporated into the matching analysis to determine which of the source spectra in the library best matches the measured activation data.

Any measurement method used to identify a neutron source spectrum relies at some point on a comparison against known sources; no identification method is definitive. Detection systems measure some physical aspect of the source, not its serial number. Many systems measure the physical aspect to determine that the source has some property, and then leave it to experts to determine which potential sources share that property. The analysis system presented here incorporates the comparison into the analysis itself. The statistical tests show which of the known library sources match the measured data the best and whether that match is a good match. If the best match still does not fit the measured data well (a very high χ^2 value), that indicates the true source is not one of the sources in the library. This is still valuable information for decision makers.

ACKNOWLEDGMENTS

This work was sponsored by the Defense Threat Reduction Agency (DTRA).

REFERENCES

1. W. E. KIMMEL, "Manufacture and Testing of an Activation Foil Package for Use in AFIDS," Masters Thesis, Air Force Institute of Technology, Wright-Patterson Air Force Base, OH, March 2005.
2. Z. W. BELL and L. A. BOATNER, "Neutron Detection via the Cherenkov Effect," *IEEE Trans. Nucl. Sci.*, **57,** 3800 (2010).
3. Z. W. BELL and L. A. BOATNER, US Patent 7,952,075, issued May 31, 2011; US Patent 7,601,965, issued October 13, 2009; US Patent 7,629,588, issued December 8, 2009.
4. J. P. HAYWARD, C. L. HOBBS, Z. W. BELL, L. A. BOATNER, R. E. JOHNSON, J. O. RAMEY, G. E. JELLSION, Jr., and C. R. LILLARD, "Characterizing the radiation response of Cherenkov glass detectors with isotopic sources," *Journal of Radioanalytical and Nuclear Chemistry*, **295**(2), 1143–1151 (2013).
5. J. P. HAYWARD, Z. W. BELL, L. A. BOATNER, C. L. HOBBS, R. E. JOHNSON, J. O. RAMEY, and G. E. JELLISON, "Simulated response of Cherenkov glass detectors to MeV photons," *Journal of Radioanalytical and Nuclear Chemistry*, **295**(2), 1321–1329 (2013).
6. T. GOORLEY, "New York City MCNP Neutron Spectra Report," LA-UR-10-06590, Sept 29, 2010.
7. Radiation Effects Research Foundation, "Reassessment of the Atomic Bomb Radiation Dosimetry for Hiroshima and Nagasaki—Dosimetry System 2002," http://www.rerf.jp/, Editors: Robert W. Young and George D. Kerr (2005).