FIRST FOUR-ISOTOPE CALIBRATION OF A BETA-GAMMA QUAD DETECTOR

Matthew W. Cooper, Derek A. Haas, James C. Hayes, Lance S. Lidey, and Brian T. Schrom

Pacific Northwest National Laboratory

Sponsored by the National Nuclear Security Administration

Award No. DE-AC05-RLO1830/PL08-MRVL-NDD05

ABSTRACT

Identification of underground nuclear detonations relies heavily on radioxenon measurements. One radioxenon detection method depends on detecting beta-gamma coincident events paired with a stable xenon measurement to determine the concentration present. Like all measurements, the beta-gamma method relies on knowing the detection efficiency for each isotope measured.

There are several methods that are commonly used to perform a detector calibration. The most common are using a National Institute of Standards and Technology (NIST) certified sealed source, a relative measurement to compare to a calibrated detector and an absolute efficiency calibration based on radioxenon gas samples. The complication in the first method is it focuses only on the gamma detectors and does not offer a solution for determining the beta efficiency. The second method listed is not similarly constrained, however it relies on another detector to have a well know efficiency calibration. The final method using actual radioxenon samples to make an absolute efficiency determination is the most desirable, but until recently it was not possible to produce all four isotopically pure radioxenons. The production, by University of Texas (UT), of isotopically pure radioxenon has allowed the beta-gamma detectors to be calibrated using the absolute efficiency method. The first four radioxenon isotope calibration will be discussed is this paper.

OBJECTIVES

The monitoring of nuclear explosions relies on a collection of detection techniques. Each technique yields a portion of the information necessary to determine when, where and what happened. While some techniques may only be relevant under specific conditions, other techniques are less dependent on environmental conditions. Detection of radioxenon effluents from a nuclear test is one of the more robust techniques and is one of the only methods available to determine whether a test is nuclear or not. However, radioxenon detection does offer its own challenges.

The challenge that will be discussed in the following paper is the calibration of the nuclear detector portion of a radioxenon system. To accurately measure radioxenon concentrations the nuclear detector must be well calibrated. The calibration can be performed in a variety of ways: from performing a calibration relative to a known calibrated detector to using National Institute of Standards and Technology (NIST) standards. However, the work presented in this paper uses an absolute calibration technique that is similar to those used by NIST to make a standard. By making the absolute measurement directly it is possible to remove additional sources of uncertainty and yield an overall better calibration. Traditional calibrations based on the absolute method relied solely on ^{133}Xe and ^{131m}Xe, however recent developments at University of Texas have led to the availability of the two short-lived isotopes ^{135}Xe and ^{133m}Xe. This paper discusses the first absolute calibration of a beta-gamma (β-γ) detector using all four radioxenon isotopes (^{135}Xe, ^{133m}Xe, ^{133}Xe and ^{131m}Xe).

RESEARCH ACCOMPLISHED

Calibration Methods

There are three typical calibration methods employed in characterizing a nuclear detector. The first and easiest method is to use a gamma standard, or NIST standard. In this method the activity of a source is accurately measured using a well-calibrated detector. The measured (often considered known) source activity is then used to determine what the detection efficiencies are for the nuclear detector of interest. This method is only as good as the calibration of the well-known detector. Another limitation is the selection of isotopes that have gamma rays of the same as or near the energy of gamma rays from expected samples. Often it is necessary to interpolate the efficiency results, which will add to the overall measurement uncertainty.

An alternative is to calibrate with a radioactive sample that has been measured using a well-characterized nuclear detector that has already been calibrated in an absolute way. This method will remove the necessity of interpolating between energies since the calibration source can be the same material expected for sample measurements, in the case of radioxenon this would be one of the four xenon isotopes of interest (^{135}Xe, ^{133m}Xe, ^{133}Xe and ^{131m}Xe). However, this method is still reliant on how well the efficiency calibration is known for the well-characterized nuclear detector. Furthermore, a new uncertainty needs to be accounted for; the understanding of uncertainties in a gas sample transfer is extremely challenging.

Due to the difficulties in the previous methods a third method has been chosen for β-γ nuclear detector (Cooper, 2005; Cooper, 2007) calibration at Pacific Northwest National Laboratory (PNNL). The third method exploits the β-γ coincident measurement to make an absolute activity measurement (NCRP 1985) and, consequently, an efficiency calibration. The absolute efficiency calibration method uses the four radioxenon isotopes (^{135}Xe, ^{133m}Xe, ^{133}Xe and ^{131m}Xe) that are to be detected as the calibration standard. The method is not reliant upon a known activity sample since it inherently determines the absolute activity but does need isotopically pure samples to obtain an accurate calibration.

β-γ Calibration Methodology

The technique can be used in a more general sense, but there are several components that need to be accounted for which will make the method extremely complex. For the β-γ application discussed in this paper there are several simplifications that can be used (Knoll 2000). For instance, the detector geometry allows for nearly 4π solid angle coverage for β-decay, meaning the sample is surrounded by the β-detector, which allows angular correlations between radiation types to be ignored. In addition, the radioxenon samples are produced in nearly radioisotopically pure form so there are no complicating radioactive interference terms.

The specific method leverages the β-γ coincidence detection to determine the total number of decays (or absolute activity) by comparison between the number of β single, γ single and β-γ coincidence detected decays.

The total activity (A_T) can be written in three different forms, one for each type of decay: β-γ, γ and β.

$$A_T = \frac{C_{\beta\gamma}}{BR_{\beta\gamma}\varepsilon_{\beta\gamma}} = \frac{C_{\beta\gamma}}{BR_{\gamma}\varepsilon_{\beta\gamma}} \quad (1)$$

$$A_T = \frac{C_{\gamma}}{BR_{\gamma}\varepsilon_{\gamma}} \quad (2)$$

$$A_T = \frac{C_{\beta}}{1-(1-\varepsilon_{\beta})\left(1-\varepsilon_{CE_i}BR_{CE_i}\right)} \quad (3)$$

Where, BR is the known branching ratio, ε is the detection efficiency, C is the number of observed decay events, and the subscript symbols γ, β and CE (conversion electron) are radiation types. It is assumed the beta branching ratios are 100% and that $\varepsilon_{\beta\gamma} = \varepsilon_{\beta} \cdot \varepsilon_{\gamma}$.

By solving (1) with respect to (2) it is possible to determine the absolute β efficiency (ε_{β}) for the particular region of interest (ROI). However, the most complex and difficult efficiency to determine is the γ efficiency. The challenge is due to the multiple decay paths that are possible. Each decay path offers another opportunity to detect γ and β or CE, which will increase the probability that any given nuclear decay is observed. These additional decay paths are taken into account by determining the probability that any one decay mechanism, and thereby a nuclear decay, is detected.

An example of the complexity of the calculation is ^{133}Xe. As can be seen in Figure 1 the primary signatures for ^{133}Xe are the ~80 keV γ-ray in coincidence with a β and the ~30 keV x-ray in coincidence with a 45 keV CE. However, there are two other decay paths that can occur in anti-coincidence with the primary. The additional decay paths result in several terms that need to be included in the calibration calculation.

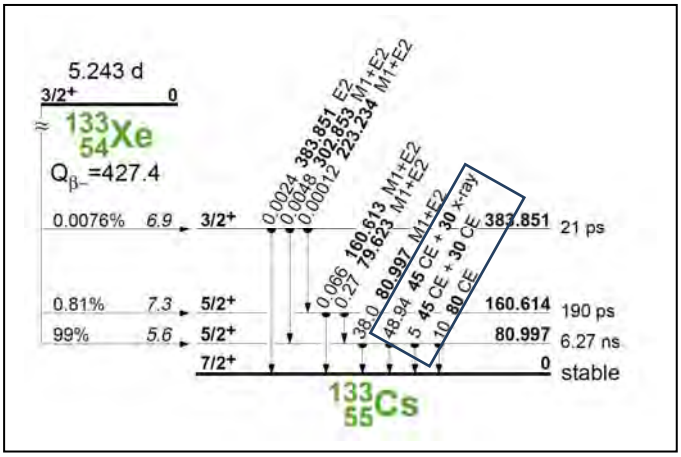

Figure 1. The γ-ray level scheme for ^{133}Xe with three additional decay paths included boxed in blue. The first number in the diagonal labels is the branching ratio, which is the probability of that particular decay occurring. The second number is the energy or approximate energy of the particular decay. For the three boxed decays the additional information gives secondary radiation signatures that are in coincidence with the decay.

Radioxenon Samples

One other critical component to the calibration of radioxenon β-γ systems is the availability of isotopically pure xenon. There are four radioxenon isotopes that are of interest both as nuclear explosion signatures and calibration sources. Until recently only two of the four have been available. Both ^{133}Xe and ^{131m}Xe have been available through standard medical isotope facilities. However, recently University of Texas has built the capability to supply ^{135}Xe (Figure 2) and ^{133m}Xe (Figure 3) in addition to the other two isotopes. The availability of all four xenon isotopes allows the absolute calibration of β-γ systems for all four isotopes. There are however several constraints to the calibration.

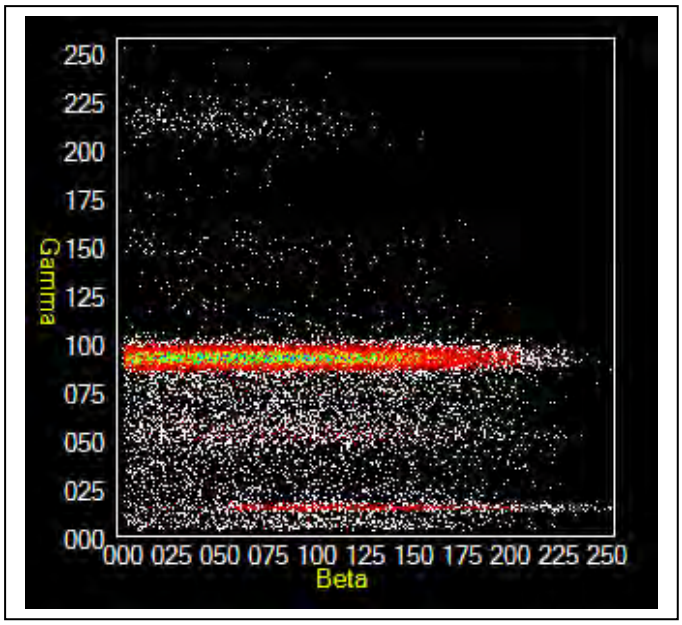

Figure 2. A β-γ coincidence plot of the ^{135}Xe calibration sample data.

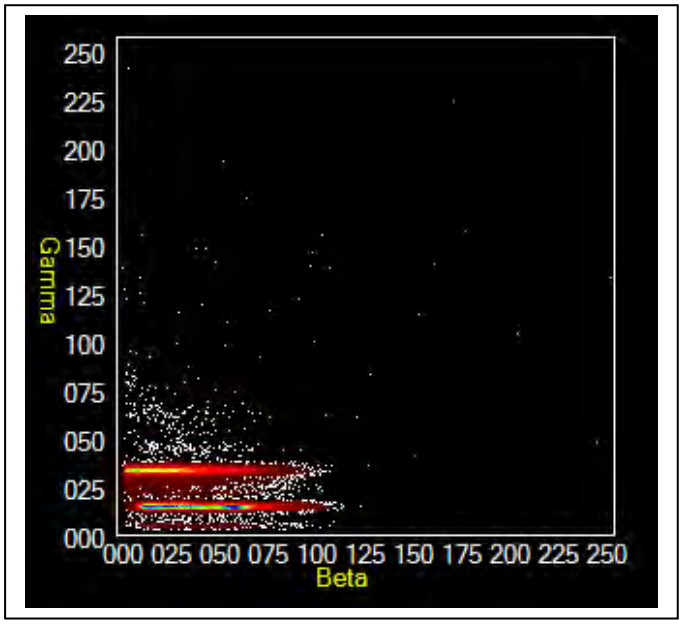

Figure 3. A β-γ coincidence plot of the ^{133m}Xe calibration sample data. The region circled in blue is the ^{133m}Xe peak, with the other horizontal peaks being ^{133}Xe.

First, there are time constraints for the calibration of the systems. Each radioxenon isotope has a different half-life, so it is important to calibrate the β-γ systems using the correct radioxenon sample order. The order progresses from the shortest half-life (^{135}Xe) isotope to the longest lived (^{131m}Xe). The order allows for the fastest calibration. In addition, due to the memory effect in the beta cell, adequate time needs to be given for each radioxenon isotope to decay away prior to the introduction of a new radioxenon isotope. The process will take on the order of a month's time to complete.

Second, the importance of a very precise low-uncertainty measurement is critical. Although the calibration method is useable for standard sample measurements because it determines the absolute activity present, standard samples typically have very low counting statistics. Consequently, it is possible to determine the b-g detection efficiency to very high level in a calibration setting and avoid additional uncertainty terms. This need to determine the detection efficiency to a high level requires high counting statistics, and therefore high activity samples. In general the uncertainty inherent in a measurement due to the counting statistics is the square root of the number of counts in the ROI. Therefore, to obtain a 1% uncertainty a minimum of 10000 net counts in the regions of interest is necessary. Additionally there will be background radiation that needs to be accounted for. This means that a shorter count time is desirable to reduce the impact of the background term. These effects drive the sample activity limits at the time of introduction into the system.

Finally, there are some small amounts of contaminants present in the samples. These contaminants are a very small fraction of the total activity, but begin to be observable in the data after several half-lives of the sample isotope of interest, meaning the samples need to be introduced into the β-γ detector relatively quickly based on the isotope half-life. For instance, ^{135}Xe has a ~9 hour half-life and has a ^{133}Xe contaminant (~5 day half-life); this means that after 2 days the ^{133}Xe will begin to impact the ^{135}Xe calibration results. The presence of contaminants primarily effects ^{135}Xe and ^{133m}Xe due to the shorter half-lives.

CONCLUSIONS AND RECOMMENDATIONS

PNNL has developed an absolute calibration method for each of the four radioxenon isotopes of interest. The calibration method will allow b-g systems to be calibrated without prior knowledge of the isotope activity or the need for cross-comparison verifications to be performed. However, the calibration method currently requires isotopically pure radioxenon samples. Fortunately, the capability to generate all four radioxenon isotopes of interest in a highly isotopically pure form has been developed at UT, allowing for a complete b-g system calibration to be performed. PNNL has performed the first absolute calibration of a b-g system using all four radioxenon isotopes.

Although this effort was completed successfully, additional effort in reducing the time between the generation and measurement of the short-lived radioxenon isotopes (in particular ^{133m}Xe) will significantly help the quality of the results. There are also additional minor sources of uncertainty that need to be addressed to further improve the calibration methodology.

ACKNOWLEDGEMENTS

The authors would like to acknowledge all of the effort over the years by multiple people over several organizations to make this work possible.

Also, thanks to Dr. Steve Biegalski for his involvement in the production of the isotopically pure radioxenon used in the b-g detector calibration.

REFERENCES

A Handbook of Radioactivity Measurements Procedures (1985). NCRP Report No. 58, 2nd edition, National Council on Radiation Protection and Measurements, Bethesda, MD, 74–93.

Knoll, G. F. (2000). *Radiation Detection and Measurement*, 3rd edition, 674–675.

Cooper M. W., A. J. Carman, J. C. Hayes, T. R. Heimbigner, C. W. Hubbard, K. E. Litke, J. I. McIntyre, S. J. Morris, M. D. Ripplinger, and R. Suarez (2005). "Improved β-γ coincidence detector for radioxenon detection," in *Proceedings of the 27th Seismic Research Review: Ground-Based Nuclear Explosion Monitoring Technologies*, LA-UR-05-6407, Vol. 2, pp. 779–786.

Cooper, M. W., J. I. McIntyre, T. W. Bowyer, A. J. Carman, J. C. Hayes, T. R. Heimbigner, C. W. Hubbard, L. S. Lidey, K. E. Litke, S. J. Morris, M. D. Ripplinger, R. Suarez, and R. C. Thompson (2007). *Nuclear Instruments and Methods. in Physics Researc. Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 579:1, 426.