LA-UR- 01-3747

Approved for public release; distribution is unlimited.

Title: Sensitivity and Accuracy Considerations for Neutron Assay of Plutonium-Contaminated Waste in Large Containers

Author(s): Sheila G. Melton, NIS-6
Robert J. Estep, NIS-6

Submitted to: To be presented at the 42nd Annual Meeting, Institute of Nuclear Material Management (INMM), Indian Wells, CA, July 15-19, 2001.

Los Alamos
NATIONAL LABORATORY

Sensitivity and Accuracy Considerations for Neutron Assay of Plutonium-Contaminated Waste in Large Containers

Sheila G. Melton, Robert J. Estep
Los Alamos National Laboratory
Los Alamos NM 87545

ABSTRACT

Since the 1970s, innovations have allowed both active and passive neutron techniques to address various safeguards and waste measurement needs in the DOE complex. Much research was focused on satisfying the 100-nCi/g detection limit for TRU waste in 208-liter drums. The emphasis on measuring drum-sized containers for disposal at WIPP has resulted in improved waste assay capability that now needs to be extended to larger containers. The desire to expedite the decontamination and decommissioning of certain DOE facilities, and the large waste encountered in that process, has prompted the need for increasingly large disposal containers. Instruments have recently been built to accommodate crates that are nearly 100 cubic feet in volume, such as a B-25 box or Standard Waste Box.

The density of hydrogen inside a waste container profoundly affects the accuracy of neutron measurements, and the metal content greatly affects sensitivity. Depending on the matrix, and especially the hydrogen content, the response of an instrument to a single point source can vary tremendously within the container. Because the density and composition of metals inside each container are unknown, the observed cosmic ray background rate varies from one container to the next, resulting in a loss of sensitivity for passive counters. In the paper we will explore the magnitude of these problems for both metal- and hydrogen-bearing matrices in a crate-sized containers.

INTRODUCTION

A measurement procedure is designed to determine the true value of some property of interest, i.e., in this case, the total amount of plutonium contained in a crate of waste. However, any measurement will produce non-identical results even repeated under completely identical conditions. Precision refers to the general reproducibility of a measurement system can be summarized by the standard deviation of the system response over a long period of time. The accuracy of a measurement refers to the overall measurement error with contributions from both random error and system bias. Thus to characterize the overall measurement accuracy, it is necessary to determine both the measurement precision and assign appropriate bounds on any source of bias in the uncertainty for the measurement results.

The overall error in the determination of the plutonium mass contained in a crate will depend on both the uncertainties in the measured response and the values used in the determination of the matrix calibration factor. As stated in a 1997 Idaho National Laboratory study on the determination of TMU for active assays of sludge waste[1],

> "...the major issue in quantifying systematic and random uncertainties involves the validity of the uniform matrix and uniform source premise used in determining the matrix calibration factor to the assay of a particular class of waste."

The question then becomes what is the magnitude of the error introduced as a result of making these two assumptions. The results presented in this paper assume a homogenous matrix and explores only the effects of source position on the measured response. For both active and passive neutron crate-sized systems, the measurement uncertainty due to non-uniform source distribution depends largely on the hydrogen density of the matrix materials.

A commonly used definition of sensitivity or minimum detectable mass in regards to nondestructive assay systems is the smallest mass that gives a signal that is three times its uncertainty. The confidence interval is chosen as 95% or 99% representing signals that are two-sigma and three-sigma above background, respectively.[2] Using this definition, sensitivities of neutron systems are quite frequently quoted as one mass value based on the measured response for a source centered in an empty chamber. Sensitivity may also be provided in grid form where efficiency is shown as a function of position within an empty crate. Although both single and positional sensitivities for the empty chamber are valid performance measures for any nondestructive assay system, they can be misleading. Most existing active and passive neutron counters are designed with 4-π detector geometry and therefore have an essentially uniform response over the volume of an empty chamber, i.e., positional variation less than 5 %. For crates containing actual waste materials, the sensitivity may vary by an order of magnitude over the volume of the crate. Sensitivity values quoted in performance specification sheets are usually calculated using a constant background assumption and a measurement period greater that the stated average assay time. Since the cosmic ray neutron background changes over time as well as with matrix material, the sensitivity in an actual waste crate may deviate significantly from the best-case scenario as shown in system performance data.

NEUTRON DETECTOR SYSTEMS - THE MEASURED RESPONSE

In general, active neutron assay consists of measuring the induced signal from the interrogation of fissile isotopes by an intense neutron source. In the waste field, the preferred active method is the differential dieaway technique (DDT). DDT uses a thermalized neutron flux from a 14-MeV generator as the interrogating source and an array of cadmium-wrapped ^{3}He proportional detectors to detect the prompt fission neutrons emitted by the fissile isotopes, ^{239}Pu, ^{241}Pu, or ^{235}U. The measured count rate is a function of both the magnitude of the interrogating flux and the total neutron detection efficiency. Since the interrogating flux and detection efficiency are both uniform throughout the volume of an empty crate, the net fissile signal also shows no positional dependence. However, absorption and moderation in matrices change the intensity of the interrogating neutron flux. The induced prompt fission neutrons are also moderated, and by a lesser degree absorbed, by the matrix materials. Thus, the number of signal neutrons that reach the detectors changes with matrix type, resulting in variation of the overall detector efficiency. Matrix effects on both the interrogating and induced-fission signals are also functions of positions within the crate.

Passive neutron assay relies on the detection of neutrons from (α,n) reactions or spontaneous fission events occurring in the desired isotope. For the passive assay of plutonium-contaminated waste where the total number of neutrons from interfering sources varies widely, coincidence counting is generally preferred. The goal of coincidence counting is to count neutrons that arise from the same fission event by using the time correlation of spontaneous fission neutrons to differentiate from random background sources. To optimize the detection of true coincidences in the presence of neutrons from random sources, the gate setting for coincidence counting of waste is usually set at 128 μs. Thus the measured response in coincidence counting (doubles rate) is proportional to the total detection efficiency squared multiplied by the gate fraction. Since coincidence counting relies on the detection of two neutrons, i.e., proportional to the detection efficiency squared, the measured response is also a function of both matrix type and source position.

While there are many possible instrument design variations, the results presented in this paper are from neutron systems that have been optimized either for the passive or active assay. The assay chambers of both crate systems are large enough to accommodate a standard waste box. For the passive assay system, more than 250-10 atmosphere ^{3}He proportional detectors are positioned in a two-row design with approximately 2 inches of polyethylene between the assay chamber and the first row of tubes. Detection efficiency is 40% for a ^{240}Pu source positioned in the center of the assay chamber. The active system uses approximately 100-4 atmosphere cadmium-shielded ^{3}He proportional detectors embedded six inches from the graphite chamber wall. Detection efficiency for a ^{240}Pu source centered in the assay chamber is 14%. Both active and passive systems are designed with 4-π detector geometry with a measured response over the volume of the chamber that is essentially uniform.

NON-UNIFORM SOURCE DISTRIBUTION

Calibration is a method to determine the relationship between a measured response and the mass of the isotope being measured. In an active or passive neutron waste assay, calibrations are performed using known plutonium standards in an empty container. Measured quantities and an empirical formulation based on either the add-a-source technique for a passive system or flux monitor data for an active are then used to correct for containers with unknown matrices. Both types of matrix corrections rely on the assumption of a uniformly distributed source throughout the container.

Assuming a constant homogeneous waste configuration, the magnitude of the spatial variation in the measured response will be a function of the size of the waste container, the density of hydrogen in the matrix, and the neutron assay method used. In general, as the size and hydrogen density of the waste increase, so does the variation in measured response. For an active neutron system, the positional variation of the net fissile signal is directed related to the variations in the interrogating flux and the total detection efficiency within the matrix. Fortunately, these two variations are directly opposed and at some hydrogen density will cancel, 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. For a passive neutron system where the desired signal is proportional to the efficiency squared, the variation in measured response only increases with hydrogen density, i.e., there is no offsetting effect. In both neutron systems, the measurement error due to spatial non-uniformly increases with matrix size until at some combination of hydrogen density and size, sources located in the matrix center can not be measured.

Table 1 shows the variation in measured response in a 675 lb. matrix with a hydrogen density of 0.0214 g cm^{-3} contained in a standard waste box. The results in Table 1 are from MCNP calculations using the specifications from an optimized passive neutron counter and data concerning combustibles waste from the Rocky Flats Plant. The relative standard deviation in the measured response over the 80 positions is 39 %. A better measure of the magnitude of the error for localized sources is the ratio of the average response to the extremes, i.e. maximum and minimum. For a passive calibration using the uniform source distribution, the measured response could be biased low by a factor of three or biased high by a factor of two.

Table 1. Variation of measured response of a passive neutron system for a 675 lb. matrix with a hydrogen density of 0.0214 g cm^{-3} contained in a waste box.

Position	Detection Efficiency-Effy (%)	Gate Fraction-GF (%)	$Effy^2$ x GF (~Measured Response)
1	30.0	63.7	5.76
2	28.2	61.4	4.88
3	22.8	58.1	3.02
:	:	:	:
:	:	:	:
80	34.2	68.8	8.00
Average	**26.9**	**60.4**	**4.60**
%RSD	**16.5**	**9.2**	**39.1**
		Ave/Max	**0.52**
		Ave/Min	**3.33**

Figure 1 shows the variation in the measured response for the same passive counter using the same hydrogen density (0.0214 g cm^{-3}) but with a maximum loading of 4000 pounds. The measured response for this matrix could be biased low by a factor of approximately 40 or biased high by a factor of 4. The same relative error magnitudes are present if we lower the matrix weight to 1400 pounds but double the hydrogen density (0.043 g cm^{-3}). For this case, the passive response could be biased low by a factor of 45 and high by a factor of 4.

Figure 1. Variation in measured response of a passive neutron system for a 4000 lb. matrix with a hydrogen density of 0.0214 g cm^{-3} contained in a waste box.

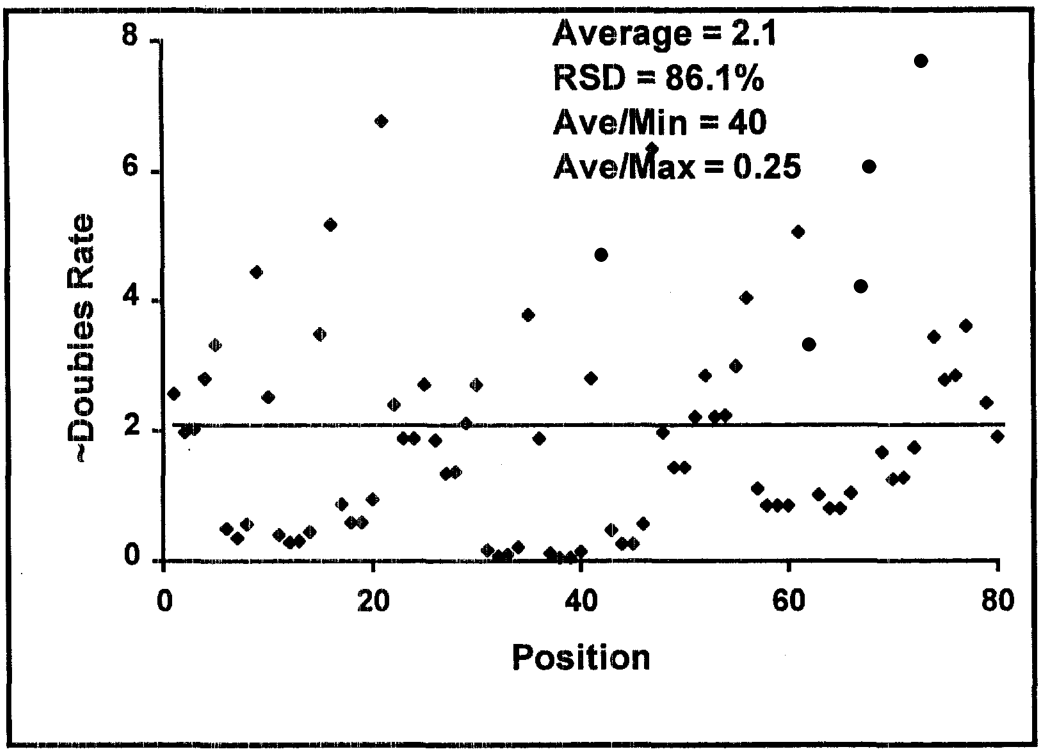

Table 2 shows the measured variation of the interrogating flux, detection efficiency, and measured response in the same 675 lb. standard waste box with a hydrogen density of 0.0214 g cm^{-3} for a neutron system optimized for the active assay. At locations near the center of hydrogenous matrices, the thermal interrogating flux is increased, thereby increasing the fission reaction rate. Neutrons resulting from fissions near the center are also moderated, thus reducing the probability that they will escape the container and be detected. At some combination of hydrogen density and depth within the matrix, the two effects of moderation will approximately cancel. Due to these opposing effects, the magnitude of the source positional error is significantly reduced in the active assay with a relative standard deviation of 11% as compared to 39% for the passive. The ratio of average to extremes in the active shows the measured response may be biased low by a factor of 1.6 or biased high by only 20%. In terms of minimizing the spatial error, clearly a hydrogen density of 0.0214 g cm^{-3} is quite close to the optimal for an active assay system of this size.

Table 2. Variation of measured response of an active neutron system for a 675 lb. matrix with a hydrogen density of 0.0214 g cm^{-3} contained in a waste box.

Position	**Detection Efficiency (%)**	**Flux Monitor Counts (~Flux)**	**Measured Response (mg ^{239}Pu)**
1	6.50	240,179	745.88
2	6.47	255,487	641.22
3	2.61	322,492	555.48
:	:	:	:
:	:	:	:
72	7.14	180,188	634.74
Average	**5.25**	**223,742**	**625.6**
%RSD	**20.2**	**26.2**	**11.1**
		Ave/Max	**0.84**
		Ave/Min	**1.58**

For typical combustible waste with low to medium absorption, the opposing effects of moderation will tend to reduce the positional variation of an active assay in comparison with the passive. In the case of non-hydrogenous matrices such as iron scrap, the measured response in the passive will only vary a few percent over the volume. The low moderating power of metals results in a depressed interrogating flux in the center of the crate relative to the outer edges. Since the detection efficiency is essentially uniform throughout the crate, the spatial variation in the measured response is directed related to the gradient of the thermal neutron flux. For a scrap metal matrix weighing approximately 2000 pounds, this results in a relative standard deviation of 22% for an active assay. The measured response in the active assay could be biased low by a factor of 2 or high by 20%.

SUMMARY

The desire to expedite the decontamination and decommissioning of certain DOE facilities such as Rocky Flats, and the large waste encountered in that process, has prompted the need for increasingly large disposal containers. There is already discussion of packaging waste in containers as large as 6' x 6' x 8' for shipment to WIPP. The potential sensitivity and accuracy problems already present in the current nominally 4' x 4' x 6' neutron based systems will only increase with container size. The above discussion did not address the issue of non-uniform matrices also account for large inaccuracies in actual waste matrices. Neutron imaging techniques optimized for large containers clearly are needed to address the spatial and heterogeneous matrix problems. Given the rising development and certification costs associated with new systems, at a minimum, optimization of new non-destructive assay systems should also include the ability to perform both passive and active assays.

ACKNOWLEDGEMENTS

This research was funded by the Mixed Waste Focus Area located at Idaho National Engineering and Environmental Laboratory in conjunction with the development of the Integrated Box Interrogation System (IBIS) with Canberra Industries.

REFERENCES

[1] L. G. Blackwood, Y. D. Harker, and T. R. Meacham, "SWEEP PAN Assay System Uncertainty Analysis: Active Mode Measurements of Solidified Aqueous Sludge Waste," Idaho report number INEEL/EXT-97-01273, (December 1997).

[2] P.M. Rinard, H.O. Menlove, and et.al., "The Nondestructive Assay of 55-gallon Drums Containing Uranium and Transuranic Waste Using Passive-Active Shuffler," Los Alamos report LA-12446-MS (November 1992).