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Nondestructive Assay Holdup Measurements with the Ortec Detective

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Abstract
Wing 4 of the Chemistry and Metallurgy Research facility at Los Alamos National Laboratory is to be downgraded from a Hazard Category 2 Nuclear Facility to a Hazard Category 3 Radiological Facility. Survey and holdup measurements are used to ensure that the total contamination levels present in the facility do not contribute enough activity to go above the Hazard Category 3 threshold quantities. Additionally, the measurement information provides an understanding of the cleanup and the equipment removal needs for the next step of decontaminating and decommissioning of the site.

INTRODUCTION

The Chemistry and Metallurgy Research (CMR) facility has been housing the research and experimental activities for analytical chemistry, plutonium and uranium chemistry, and metallurgy since the start of the Los Alamos National Laboratory. It is currently being replaced by the new Chemistry and Metallurgy Research Replacement facilities. As a result, the CMR is gradually closing and/or downgrading to a nonnuclear facility.

In 2008, the Safeguards Science and Technology group, N-1, was assigned the task of doing survey and holdup measurements of Wing 4 of the CMR. The goal of the measurements is to provide defensible measurement data for Wing 4 of the CMR Building to be downgraded from a Hazard Category 2 Nuclear Facility to below a Hazard Category 3 Radiological Facility. In addition, the measurement information would provide an understanding of the cleanup and the equipment removal needs for the next step of decontaminating and decommissioning the site.

The large area/volume of the site and the high intensity of the high-energy gamma rays of thorium, either from the background or the contaminated objects in the measured room or the adjacent rooms, present some challenges in the holdup measurements. Typical holdup techniques of point source, line, or area measurement [1] do not work well. In order to speed up the measurement time and to accurately account for all the isotopes present in the facility, we used a new technique that we tentatively named "Room Holdup Measurement" to do holdup measurements of the site. This technique uses the portable, electric-cooled high-purity germanium detectors from Ortec (the Detectives) to measure the activities of the isotopes.

MEASUREMENTS

We used two Detectives to acquire data. The Detective is designed to be used mainly as a nuclide identifier. We used them for a very short time at the beginning of the measurement campaign to identify the possible isotopes. For all of the later measurements, we used them as simple data-acquisition systems.

Most of the measurements were done without any collimator. For some measurements, we used the 4-mm tungsten shield to reduce the background from the surroundings in order to better measure the activity at a specific location. One or two measurements were made for a very small room, about 2 m on a side. For larger rooms, many measurements were required for accurate determination of the activities. For some very large rooms, many tens of measurements were made, with well over 100 spectra for the largest room. Each measurement was about 10 minutes long, and many were significantly longer, with some overnight and weekend measurements.

ANALYSIS

The following sections describe the different components of the analysis—isotopes used in the analysis, room background subtraction, angular efficiency calibration of the detector, and activity calculation.

Isotope Energies Used in the Analysis

In order to know which isotopes to concentrate the analysis on, several peaks were searched for in all the spectra. The search peaks are listed in Table 1. Because ^{228}Th is the progeny of ^{232}Th, all the gamma rays emitted in the decay chain of ^{228}Th can also come from the decay chain of ^{232}Th. There is one intense gamma ray, 911 keV, from the decay of ^{232}Th that is not present in the decay chain of ^{228}Th. Therefore, the 911 keV peak is used to determine the activity of ^{232}Th, and the 239, 583, and 2,615 keV peaks are used to determine the activity of ^{228}Th plus ^{232}Th. The activity of ^{228}Th is then obtained by subtracting the latter from the former.

Table 1. Gamma Rays Used in the Data Analysis

Gamma-Ray Energy (keV)	Isotope
186	^{235}U
239	^{228,232}Th
352	^{230}Th
414	^{239}Pu
583	^{228,232}Th
609	^{230}Th
662	^{241}Am, ^{137}Cs
722	^{241}Am
911	^{232}Th
1,461	^{40}K
2,615	^{228,232}Th

The thorium and uranium activities are calculated on a room-by-room basis. Given the low count rates for ^{239}Pu and ^{241}Am, these activities are calculated on a floor-by-floor basis.

Background Subtraction

The measurements show a significant amount of thorium (^{228}Th, ^{230}Th, and ^{232}Th) in the basement and other rooms of Wing 4. These isotopes are also present in the soil and concrete used to build Wing 4. It is therefore important to properly subtract the background gamma rays in order to correctly determine the amount of these isotopes present from processing activities within Wing 4 rather than those that occur naturally in the building structure itself.

To subtract the background from an assay spectrum, a background spectrum would need to be measured in an area where no thorium processing has occurred. This background spectrum would need to be normalized to the assay spectrum so that the proper amount of thorium background is subtracted out of the assay spectrum. The two spectra would need to be normalized to either (1) the spectra live times or (2) the counts in specific peaks of ^{230}Th, ^{232}Th, or ^{40}K.

The live-time normalization is correct if the measurement conditions are the same for the background and the assay spectra. For a detector that has isotropically uniform efficiency, these conditions can be easily achieved. However, it is difficult to achieve identical measurement conditions with the Detective because of its nonuniform efficiency profile. We tried this live-time normalization subtraction method and found that most of the time, the 911 keV peak from ^{232}Th and the 1,461 peak from ^{40}K had the same trend after the normalization—both positive or both negative. This means that it oversubtracts in some spectra while undersubtracting in others.

Because the source of thorium is from both the building materials and the processing activities in Wing 4, the spectra cannot be normalized using the thorium peaks. Therefore, the counts in the1,461 keV peak are used to normalize the background spectrum to the assay spectrum. The assumption in this normalization is that as the ^{40}K increases/decreases in a spectrum, the background thorium levels will also scale by the same fraction. Once the background is normalized, it can be used to remove the thorium contribution caused by building materials from the assay spectrum.

Attenuation Corrections

From the results of the 239, 583, and 2,615 keV peaks from ^{228,232}Th decay and of the 352 and 609 keV peaks from ^{230}Th decay, it is possible to correct for gamma-ray absorption resulting from self and external attenuation. Because the chemical and physical configurations of the isotopes are not known, correction for self-absorption cannot be made. Given that the material is in the form of holdup, the self-absorption effects should be small. Therefore, the assumption is that all the absorption comes from an external absorber, and the count rates are corrected using the factors for an external absorber (i.e., the inverse of the transmission). The external absorber will likely be concrete or steel. The absorption coefficients for these materials are similar; therefore, the coefficient for iron is used for all the external attenuation corrections made in this work.

The 2615 keV gamma ray, with its very high energy, can pass through thick layers of absorber to reach the detector. There is a 24% probability that its gamma rays will pass through a 6" concrete wall. Similarly, there is a 5% and a 1% chance for the 583 and 239 keV peaks, respectively, to pass through the same wall. Because there is so much thorium in the facility, there will be many 2,615 keV gamma rays entering the detector from the other rooms around the measured room. It is therefore not a good idea to use this peak for attenuation corrections. Therefore, the pair of 239 and 583 keV peaks of ^{228,232}Th and the pair of 352 and 609 keV peaks for ^{230}Th are used to correct for external absorbers.

Activity Calculation

A reasonable way to analyze the data is to assume that all the activities of the isotopes are distributed in some way in the ductwork or on the wall, floor, and ceiling of a room. The total activity is then determined by integration over all the surfaces. This integration task is difficult, but it can be simplified by assuming that the activities of the isotopes are clumped into point sources, and those point sources are distributed in some way in the ductwork and on the surfaces of the room. The integration over all the surfaces now becomes a summation of discrete point sources and can be easily obtained.

The equation for the total activity of an isotope in a room is then

$$A = \frac{R}{Br} = \frac{1}{Br}\sum_{j} R_j = \frac{1}{Br}\sum_{i,j} R_{i,j} r_{i,j}^2 / \varepsilon_\theta, \qquad (1)$$

where R is the rate of the gamma ray emitted by the isotope in the room,
Br is the branching ratio of the peak,
i denotes the detector positions of different measurements,
j denotes the assumed point source positions,
R_j is the rate of the gamma ray emitted by the point source at position j,
$R_{i,j}$ is the peak rate in the detector at position i from source j,
$r_{i,j}$ is the distance between the detector at position i and source j, and
ε_θ is the detector angular efficiency per unit area.

Equation 1 looks somewhat complicated. To simplify the math, the analysis can be performed in a different way. For each measurement,

$$R_i = \sum_j R_{i,j} = \sum_j R_j \varepsilon_\theta / r_{i,j}^2, \quad (2)$$

where R_i is the measured rate of the gamma ray i in the spectrum. For n number of measurements in the room, there will be n Equation 2s that need to be solved simultaneously to obtain results.

Equation 1 or 2 can be solved if the number of assumed point sources in the room is less than or equal to the number of measurements made in the room. That is, the number of unknowns (point source activities) must be equal to or smaller than the number of equations (the peak rate of a gamma ray in a spectrum) for it to work. There is one exception to this rule—if the activity of a point source becomes negative, which is a nonphysical result. For these sources, the activities are set to zero, which allows other point sources or other variables (such as the positions of the sources) to be added to the calculation.

Angular Efficiency Calculation

The angular efficiencies of the Detectives were measured using a small ^{228}Th source, which has three major gamma rays at 238.6, 583.2, and 2614.5 keV. From the efficiencies of these three gamma rays, the efficiency of any other gamma rays can be calculated from about 150 keV up to 3 MeV by interpolation or extrapolation. For each measurement, the source was set at a distance of 20 cm, if possible, from the center of the detector crystal. For some measurements, such as from behind the detector, a larger distance (such as 30 or 35 cm) was used because a distance of 20 cm would not be possible because of the physical shape of the Detective. The angular efficiency was measured at 15-degree intervals along the horizontal, vertical, and diagonal planes with respect to the horizontally sitting detector. Figure 1 shows the absolute efficiencies at three energies of an unshielded Detective (Detective 258) renormalized at a distance of 1 m on the

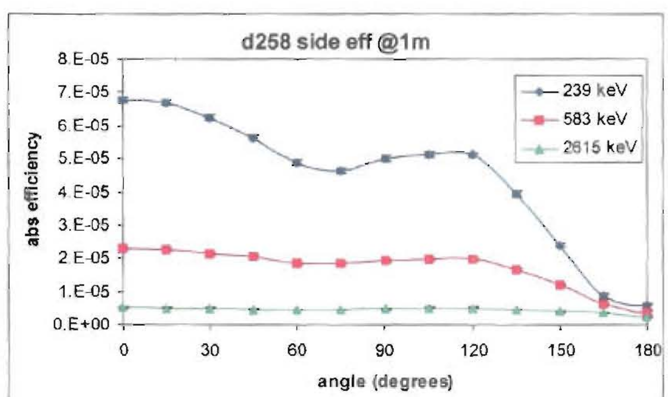

Figure 1. Absolute efficiency of the Detective 258 at 1 m distance measured from the side of the detector.

horizontal plane passing through the detector axis.

The absolute efficiency of the 2,615 keV peak is small and does not show well in the figure. In order to see the change in efficiency of this peak, the data are replotted in Figure 2. This figure shows the relative efficiencies normalized to 0 degrees at three energies.

For the efficiencies along the other planes, because of the symmetry of the detector head at angles smaller than 120 degrees, the efficiencies are the same for all the planes at angles of 120 degrees or less. For angles greater than 120 degrees, Figure 3 shows the comparison of the efficiencies.

Because many measurements at the CMR were done with the detector pointing upward with no recorded information on the other axes (top and side of the detector), the calculations were simplified by taking the average of the efficiencies of all the planes and using the average efficiency in all the calculations.

Figure 4 shows the average absolute efficiencies of three energies for the Detective 258 at 1 m distance from the source at various angles with respect to the detector axis. These efficiencies will be used in all of the activity calculations in this report. The same measurements and calculations were repeated with the tungsten shield installed on the detector and with the second high-purity germanium detector—the Detective 252—with and without the tungsten shield.

Figure 5 shows the polar plot of the relative efficiencies for the Detective 258 at three energies. This type of plot makes it easy to visualize the magnitude of the efficiency at different angles.

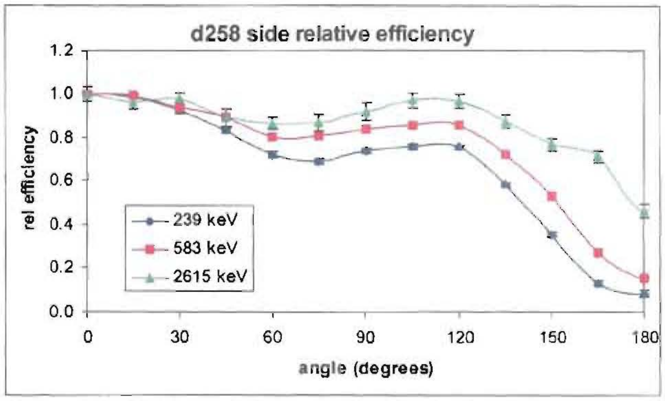

Figure 2. Relative efficiency of the Detective 258 measured from the side of the detector.

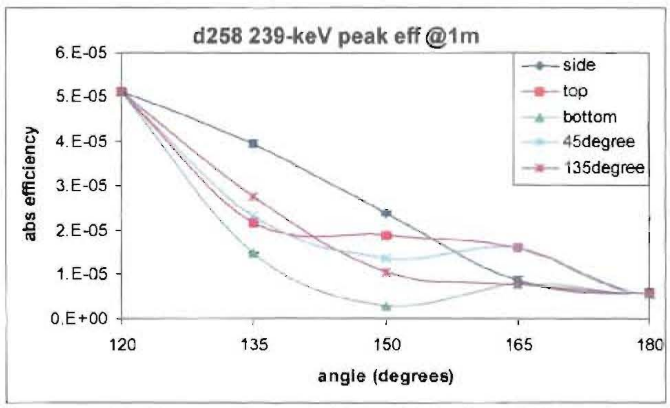

Figure 3. Relative efficiency of the Detective 258 at 239 keV at 1 m distance measured along various planes bisecting the detector crystal through its axis.

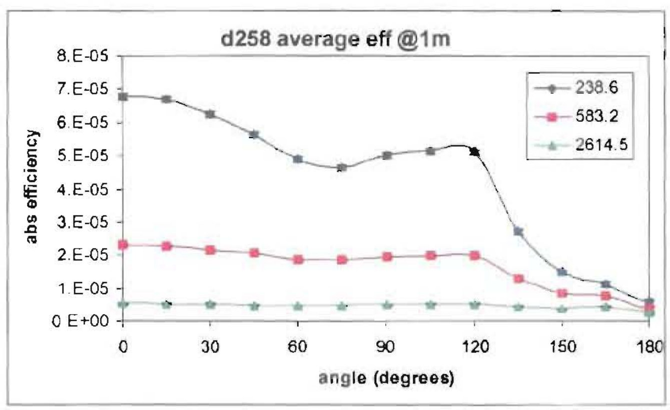

Figure 4. Average absolute efficiency of the Detective 258 at 1 m distance.

Proof of Principle—Example Calculation

As an example, the 186 keV peak activities in Room 4066 were calculated. This peak was chosen because it is the lowest energy peak measured in the spectra. As seen in Figure 4 and Figure 5, the angular efficiency of the detector varies the most for the lowest energy peak. In the following discussion, the analysis technique is shown to be valid for low-energy gamma rays. The analysis is equally valid for higher-energy gamma rays because they are less subject to attenuation effects (i.e., smaller corrections), resulting in a more uniform detector response.

The 186 keV peak activities in Room 4066 were calculated using several different assumed source configurations. Figure 6 shows the diagram of Room 4066 with the detector positions and the source positions for some of the configurations.

The dimensions of the room are about 7.4 m, 5.6 m, and 3.2 m in the x, y, and z directions. The circles (black) represent the detector positions. For these measurements, the detector was pointing upward in the z direction. The detector crystal was 1.25 m above the floor. The squares (red) represent the source positions for some of the configurations calculated.

Below are the descriptions of the configurations used in the calculations:

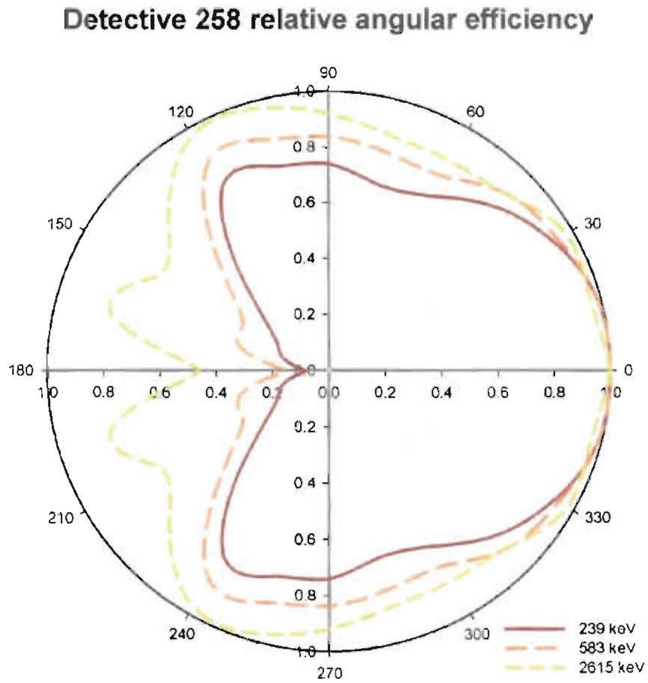

Figure 5. Average relative efficiency of the Detective 258.

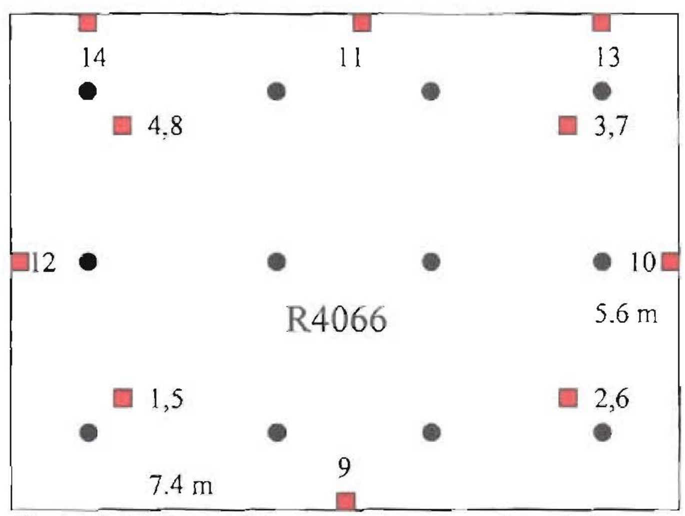

Figure 6. Diagram of the measurements and analysis configurations of Room 4066.

a. The 12 point sources were assumed to be on the ceiling directly above the detector positions.

b. The 12 point sources were assumed to be on the floor directly below the detector positions.

c. The two configurations *a* and *b* above are somewhat extreme. A more reasonable configuration would have the sources distributed somewhat evenly throughout all the surfaces of the room. Configuration *c* has the 12 sources, numbers 1–12, shown in

Figure 6, placed at the square positions. Positions 1–4 are located on the ceiling, positions 5–8 are located on the floor, and positions 9–12 are placed on the four walls at the midpoints of the walls—1.6 m above the floor.

d. After the analysis of configuration *c* above, the sources appear to concentrate near positions 1, 3, 11, and 12. Two more sources were added to the analysis at positions 13 and 14 (also 1.6 m in the z direction). Note that this configuration now has 14 source positions and only 12 measurement positions, but it still works because some of the sources were determined to have zero activity.

e. Reverted back to the 12 source positions of configuration *c*, but the sources at positions 3 and 11 are allowed to vary their positions (#3 varies in the x, y directions, and #11 varies in the x, z directions).

f. In addition to configuration *e* above, the sources at positions 1 and 12 were allowed to vary their positions (#1 varies in the x, y directions, and #12 varies in the y, z directions).

Table 2 shows the 186 keV gamma rate results. The statistical uncertainties of all these results are 20%. These results do not include the correction for self-absorption and absorption resulting from external shielding.

Table 2. Results of the Calculations of Various Source Configurations

Configuration	Gamma Rate
a. 12 sources on ceiling directly above the detector positions	68,766
b. 12 sources on floor directly below the detector positions	68,196
c. 4 sources on ceiling, 4 on floor, and 4 on wall	60,243
d. 4 sources on ceiling, 4 on floor, and 6 on wall	50,169
e. Configuration c with positions of 2 sources on upper right corner free	47,745
f. Configuration e with additional positions of 2 sources on lower left corner free	47,328

The results are largest at the extreme configurations—*a* and *b*. The reason for the large or overestimated results is a bad estimation of the source locations. As the configuration of the sources becomes closer to the real distribution, the gamma rate becomes smaller. The results representative of the true distribution are the last two, where the locations of the sources are allowed to vary. The analysis of these configurations is also more complicated and may not always be possible because allowing some sources to vary their positions will increase the number of parameters (or unknowns), and the total unknowns may exceed the number of measurements in the room. Consequently, the number of sources used in the analysis will be the same as the number of measurements in the room—as in configuration *c* above. The result of this analysis will be a small overestimate. Note that the purpose of this holdup measurement is not to accurately determine the activity of an isotope but to ensure that the total contamination levels present in the facility are below the Hazard-Category-3 threshold quantities; therefore, the slight overestimation of the activity does not invalidate our final conclusion.

The ^{235}U activity is obtained by dividing the total gamma rate of 60,243 γ/s by the 186 keV peak branching ratio (BR) of 0.574, which will then give a value of 1.05e5 Bq, or 2.84 μCi. This activity has not been corrected for the attenuation because of the external absorber. The attenuation correction is done using the activities of the major peaks from $^{228,232}Th$ decay and ^{230}Th decay.

The equation for the absorber thickness is

$$x = -\frac{\ln(T_2/T_1)}{\mu_2 - \mu_1} = -\frac{\ln(A_2/A_1)}{\mu_2 - \mu_1}, \qquad (3)$$

where x is the absorber thickness,
T_i is the transmission probability of the gamma ray i passing through the absorber,
A_i is the activity of the isotope measured by the gamma ray i using Eq. 1,
μ_i is the attenuation coefficient of the absorber at the energy of the gamma ray i,
and gamma rays 1 and 2 are from the same isotope.

Table 3 shows the gamma rate results for the major peaks of $^{228,232}Th$ decay and ^{230}Th decay for the configuration c and the iron absorber thickness calculated from those results.

Table 3. Gamma Rate Results for the $^{228,232}Th$ and ^{230}Th peaks and the Iron-Absorber Thickness Calculated from those Results

Isotope	γ (keV)	μ(cm²/g)	BR	R (1/s)	δR (1/s)	Act. (Bq)	x(g/cm²)	δx(g/cm²)
$^{228,232}Th$	238.6	0.121132	0.435	312,102	23,572	717,476	10.725	2.195
	583.2	0.07664	0.306	353,810	21,913	1,156,241		
^{230}Th	352	0.097325	0.358	134,825	16,799	376,606	7.879	6.943
	609.3	0.075107	0.448	200,999	18,283	448,659		
Average							**10.466**	**2.093**

It is noteworthy to mention that the pair of 238.6 and 2,615 keV peaks of $^{228,232}Th$ would give an absorber thickness of 16.025 ± 0.983 g/cm². The error from this pair is more than a factor of 2 smaller than that of the 238.6 and 583.2 keV pair of peaks. However, as mentioned earlier, there is a significant probability for the 2,615 keV gamma ray from the adjacent rooms to enter the detector, and this will result in an overestimation of the absorber thickness. Therefore, the 2,615 keV gamma ray is not used in either the activity calculation of the $^{228,232}Th$ activity or the attenuation correction.

After the absorber thickness is obtained, it is then used to correct for the attenuation of all the peaks of all the isotopes (see Table 1). For the record, applying this correction to the 186 keV peak of ^{235}U (with an attenuation coefficient of 0.1445 cm²/g) would result in the final activity of 12.9 ± 4.6 μCi.

CONCLUSION

We were assigned the task of doing survey and holdup measurements of Wing 4 of the CMR in order to provide data for the downgrading of the facility from a Hazard Category 2 Nuclear Facility to a Hazard Category 3 Radiological Facility. There were several factors, such as a large high-energy gamma-ray background, a large area, and an unknown physical configuration of the nuclear material and absorber that excluded the use of the typical holdup techniques of point source, line, or area measurement. We therefore invented a new technique to do holdup measurements of the rooms. This technique is simple, fast, works well, and gives reasonable results.

The final results for the entire Wing 4 were found to be sufficiently below the Hazard Category 3. This would make it possible to complete our objective of downgrading Wing 4. We are indebted to the CMR personnel, especially George Clines, and all the others who assisted us with the measurements.

Reference

1. Phyllis A. Russo, "Gamma-Ray Measurements of Holdup Plant-Wide: Application Guide for Portable, Generalized Approach," Los Alamos National Laboratory report LA-14206 (2005).