QUALIFYING THE ZEUS SYSTEM FOR VERIFICATION OF GIC ROOM TRASH FROM RADIATION CONTROLLED AREAS AT LANL

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ABSTRACT

Los Alamos National Laboratory (LANL) radiological facilities produce low-density room trash that, in many cases, is not contaminated with radioactivity. It has been estimated that 50 to 90% of low-density room trash is free of radioactive contamination and eligible for inclusion in LANL's Green is Clean (GIC) program. The GIC program is a verification program for non-regulated waste from radiation controlled areas (RCAs) that has been actively segregated as nonradioactive through the use of the waste generator's acceptable knowledge (AK).

ZEUS is the third in a set of evolving, highly sensitive systems that are optimized to detect very small quantities of common LANL radionuclides, especially isotopes of plutonium, americium, and uranium. ZEUS is designed to be particularly sensitive to the detection of very low-energy x-rays (13-21 keV) as well as low-energy gamma rays. ZEUS measures bagged waste in a 31 cm diameter by 80 cm tall circular cylinder made of a thin wall of cardboard. ZEUS employs a vertical array of four shielded Phoswich scintillation detectors placed 1.27 cm from the rotating cylinder of waste.

The ZEUS system sensitivity has been evaluated for the following matrices: paper, tyvek, and latex gloves. Detection limits for the 60 keV gamma of Am-241 are quite similar regardless of the low-density matrix material (nominally 11 Bq total activity). However, detection limits for the low-energy x-rays of Pu-239 vary substantially with different matrix materials. Detection limits for 4 kg of paper matrix are roughly 71 percent higher than for the same amount of tyvek matrix (253 Bq vs. 148 Bq). The chemical makeup of paper has some higher density atoms (e.g., oxygen) compared to a pure hydrocarbon like tyvek that cause this discrepancy at such low energies. The unique chemical composition of matrix materials has a potentially large effect on detection sensitivity at such low-energies. Therefore, we also developed a simple method to determine the acceptability of new materials for the GIC program without undergoing lengthy calibration tests in a mockup container.

INTRODUCTION

The Green-Is-Clean (GIC) program at the Los Alamos National Laboratory (the Laboratory) allows waste generators to use process knowledge to segregate clean (i.e., non-radioactive) wastes which are generated in radiologically controlled areas from radioactive wastes. GIC wastes are low-density materials (i.e., paper and plastics) that are ultimately disposed in an unregulated public landfill as opposed to the Laboratory's radioactive waste landfill. An important step in the GIC program is the screening process by which clean wastes are verified to be free of added radioactivity through appropriate high-sensitivity measurements. The ultimate sensitivity of a radiation detection system is dependent upon a variety of factors, including: background count rates, detector efficiency, source-to-detector geometry, and matrix attenuation effects.

In October 1997 the Waste Assay for Nonradioactive Disposal (WAND) system was approved for use in verification measurements [1]. Shortly after that, the High Efficiency Radiation Counter for Ultimate Low Emission Sensitivity(HERCULES) system was also approved for verification measurements in July 1998 [4]. Currently FWO-SWO has been developing a third-generation upgrade to these systems that is named ZEUS. A fundamental design change was implemented to improve ZEUS' detection sensitivity for photon-emitting radionuclides compared to HERCULES. The cylinder that contains the GIC waste was reduced in size from a 45.7 cm diameter to a 30.5 cm diameter. To compensate for the smaller diameter the cylinder height was increased by 20 cm (from 57 cm to 77 cm). Subsequently, a fourth detector was added to the top of the vertical array to inspect the waste in the upper 20 cm. The changes in the dimensions are such that the new cylinder still has enough volume to fit the contents of a typical GIC container (a 30.5 cm x 30.5 cm x 61 cm box). This report describes the system design as well as the results of FWO-SWO's technical evaluation of ZEUS' sensitivity as a GIC waste verification system.

SYSTEM DESCRIPTION

ZEUS (Fig. 1) is a prototype system consisting of a vertical array of four Phoswich detectors positioned adjacent to a rotating turntable in a well-shielded detection chamber. Low-density GIC wastes are placed inside a thin, but rigid, cardboard cylinder on the turntable. Wastes can be measured for any preset count time, depending on detection sensitivity requirements. However, the default measurement time used for this technical evaluation was 1,000 seconds.

Fig. 1 The ZEUS system detection chamber and measurement cylinder

The large area (127 cm^2) Phoswich detectors consist of a thin NaI front crystal (3 mm thick) that is optically coupled to a thick CsI back crystal (50 mm thick). The crystals are set in an oxygen-free high-conductivity copper housing with a very thin (0.025mm) aluminum entrance window. The scintillation properties of the two crystals are different enough to allow the system's electronic components to identify the origin of any pulse. The custom multiplexer uses decay-time discrimination to separate signals into two distinct spectra for analysis. Therefore, a single Phoswich detector provides the information of two detectors at one time: 1) a spectra of low-energy events (NaI) and 2) another of higher energy events (CsI). Furthermore, we employ coincidence rejection of preamplifier pulses that exhibit simultaneous scintillation characteristics of both crystals. Rejection of coincidence pulses results in a reduction in background radiation rates in the low-energy spectra since such events usually occur from higher energy photons that have a scattering interaction in the NaI crystal. The net effect of coincidence rejection is that background signals in the NaI crystal are reduced by about a factor of four.

The ZEUS detection chamber has dimensions of 107 cm x 51 cm x 91 cm (L x W x H). GIC wastes are placed in a thin-walled cardboard cylinder that rests on a rotating turntable (12 rotations/min). The detector array is located 1.27 cm (0.5 inches) adjacent to the rotating cylinder with the detectors evenly spaced in the vertical plane. A sliding door on the top of the detection chamber allows for access to GIC wastes in the waste cylinder. The chamber walls are filled with two inches of lead shielding and are lined on the interior with 0.8 mm (1/32nd inch) each of cadmium and copper sheets. Graded shielding with Cd and Cu is a well-known technique to reduce the backscattering of fluorescent lead x-rays into the low-energy portion of the NaI spectra.

METHODS

Based on previous experience, we knew that the detection sensitivity for a single point source of contamination would vary with the position of the source in the container and with the type and quantity of attenuating material in the waste matrix. A single point source of contamination is considered to be the most realistic scenario for measurement trials. This is because GIC wastes are segregated using waste generator process knowledge and we expect that only a very small fraction of individual waste items will have any added contamination whatsoever.

A weapon's grade Pu source with known amounts of Pu-239 and Am-241 was placed at different positions with respect to the radial and vertical axes of the drum. The Pu-239 x-ray equivalent activity of the source was 1.54E+05 Bq and the Am-241 activity was 3.05E+03 Bq. The following positions were chosen for the radial axis with respect to their distance from the center of the drum: 0, 3.81, 7.62, 11.43, and 14.0 cm (Note: 14.0 cm represents a source placed nearly against the side of the cylinder wall). Five vertical positions were chosen: 0, 10, 20, 30, and 40 cm (Note: 40 cm corresponds to the vertical center of the cylinder). Only positions in the bottom half of the detection chamber were examined since it is reasonable to expect that radiation signals from the top half of a uniformly distributed matrix would mirror the results from positions in the bottom half. However, we believe that there were some unavoidable compaction effects in the bottom half of the latex glove matrix. Therefore, the calculated system efficiency for this matrix and the associated detection limits are probably somewhat conservative relative to the 'true' values.

Three common low-density waste matrices were chosen as the materials for our mockup containers: 1) 4 kg of shredded paper, 2) 4 kg of tyvek coveralls, and 3) 8 kg of latex gloves. In each case the matrix fully filled our test cylinder (0.056 m^3 volume) without undo compaction. The source was measured in the rotating cylinder for count times that varied from 600 sec to 2000 sec. We always attempted to count long enough so that each key region of interest (ROI) accumulated at least 10,000 counts. The exceptions to this occurred mostly in the more dense latex glove matrix, particularly when the source was placed in the center of the measurement cylinder. However, the ROI counts were always greater than 4,000 which still yields a reasonably low statistical error. We evaluated the following two ROIs to determine the detection limits for our source: 1) the low-energy x-ray region from 13-21 keV for Pu-239 and 2) the 59.54 keV region for Am-241.

The ZEUS system will be evaluating count rates in 16 ROIs for each measurement it performs – four ROIs in each of four detectors. It is standard practice to allow a 5% false positive rate while evaluating individual data points. However, if we allowed each ROI to have a 5% false positive rate, then we would have at least one false positive result in 80% of our measurements – an unacceptably high rate for routine operations. Therefore, we increased our critical level (L_C) so that we would only incur a total false positive rate of 5% across all ROIs (Note: The critical level is defined as the count rate at which we claim that activity above background is present). The false negative rate (Type II errors) for the 16 ROIs can remain at 5% since this is an *a priori* judgment that will not affect our operations. Therefore, the detection limit (L_D) is calculated according to the equations below [2]:

$$L_D = L_C + k_\beta \sigma_b \quad \text{(Eq. 1)}$$

Where σ_b is the standard deviation of the background counts. The critical level that produces a 5% false positive rate is:

$$L_C = 2.74\sigma_b \quad \text{(Eq. 2)}$$

Since we are accepting a 5% false negative rate, we are setting k_β to equal 1.645. Substituting these values in Eq. 1 the detection limit becomes:

$$L_D = 4.385\sigma_b + 3 \quad \text{(Eq. 3)}$$

RESULTS

The first step in evaluating the detection sensitivity of the ZEUS system was to determine the background radiation rates in various energy regions of interest (ROIs). It is necessary to know the background rates of both low-energy ROIs in the NaI spectra as well as the higher energy ROIs in the CsI spectra. A number of 50,000 sec background measurements were performed during overnight counts. Our previous experience with HERCULES indicated that background rates can vary with the total mass of low-density material present in the detection chamber. This occurs because low-density (i.e., low Z) materials will preferentially scatter photon radiation instead of absorbing it. In an empty chamber most high-energy photons will travel through without an interaction until they encounter lead shielding on the opposite wall. However, the probability of a Compton scattering interaction increases significantly when low-density material is placed in a photon's path. Therefore, we carefully examined variations in background rates with the total mass of material in the waste cylinder.

Average background rates for the four ZEUS detectors are presented in Table I. The results are somewhat curious in that background rates increased significantly with increases in mass in two of the four ROIs, yet showed virtually no change in the other two ROIs. The two ROI's that exhibited significant rate increases with mass are both in the low-energy NaI region of the spectrum: 59.5 keV and 92.6 keV. Given this, one would also expect a similar increase in the other low-energy ROI – the x-rays from 13-21 keV. However, this was not observed. We are guessing that additional photons were probably initially scattered into this very low-energy region. However, at these very low-energies it is possible that most of them were ultimately absorbed in the matrix through photoelectric interactions before they had the opportunity to interact with the detector's NaI crystals.

Table I Average Background Radiation Rates in ZEUS Detectors

Energy Region	Rate (cps) in Empty Chamber	Rate (cps) with 8 kg Paper Matrix	Percent Difference	Statistically Significant?
13-21 keV	0.2337	0.2360	0.98	No
59.5 keV	0.1763	0.2178	23.54	Yes
92.6 keV	0.2160	0.2841	31.53	Yes
661.7 keV[a]	1.8475	1.9187	3.85	No

[a] Indicates an ROI in the CsI portion of the spectra.

The detection sensitivity of ZEUS to the low-energy x-ray emissions of Pu-238 and Pu-239 is considered the system’s most critical performance measure. This is because the low x-ray yield per decay (4.65% for Pu-239 and 10.7% for Pu-238) coupled with the very low-energy of the emissions make them the most challenging photon signal to detect of all the potential radionuclides in LANL’s GIC waste streams (Note: Although it is a common radionuclide in LANL’s wastes, tritium is specifically excluded from the GIC program). Furthermore, plutonium isotopes are highly radiotoxic and usually have the most stringent regulatory limits for free release. The ANSI/HPS standard, ANSI/HPS N13.12, Surface and Volume Radioactivity Standards for Clearance, proposes a volume limit of 0.11 Bq/g (3.0 pCi/g) [3]. Therefore, our initial efforts have focused on the performance of the ZEUS system for detecting Pu x-rays and Am-241. The Am-241 is included because it is nearly always present in weapon’s grade plutonium as the decay product of Pu-241 – and – because it’s abundant 59.5 keV gamma-ray often makes it the most readily detectable signal in the weapon’s grade Pu mixture.

Table II presents the MDAs and MDCs (minimum detectable concentrations) for Pu-238, Pu-239, and Am-241 for a uniformly distributed contamination source. In addition, Table III presents those same results for the worst-case location of point source contamination (i.e., the center-bottom location). All values are for a 1,000 sec count time.

Table II. MDAs and MDCs for Pu-238, Pu-239, and Am-241: Uniform distribution

	Pu-238		**Pu-239**		**Am-241**	
	MDA (Bq)	**MDC (Bq/g)**	**MDA (Bq)**	**MDC (Bq/g)**	**MDA (Bq)**	**MDC (Bq/g)**
4 kg Paper	1.10E+02	2.75E-02	2.53E+02	6.33E-02	1.10E+01	2.75E-03
4 kg Tyvek	6.44E+01	1.61E-02	1.48E+02	3.70E-02	1.07E+01	2.66E-03
8 kg Latex	1.28E+02	1.60E-02	2.96E+02	3.70E-02	1.11E+01	1.39E-03

Table III. MDAs and MDCs for Pu-238, Pu-239, and Am-241: Worst case location

	Pu-238		**Pu-239**		**Am-241**	
	MDA (Bq)	**MDC (Bq/g)**	**MDA (Bq)**	**MDC (Bq/g)**	**MDA (Bq)**	**MDC (Bq/g)**
4 kg Paper	3.65E+02	9.13E-02	8.40E+02	2.10E-01	1.90E+01	4.75E-03
4 kg Tyvek	1.85E+02	4.63E-02	4.26E+02	1.07E-01	1.86E+01	4.65E-03
8 kg Latex	1.62E+03	2.03E-01	3.74E+03	4.67E-01	2.13E+01	2.66E-03

When considering the results for uniformly distributed contamination, the MDCs for all matrices and nuclides are below the ANSI/HPS N13.12 standard of 0.11 Bq/g. However, in the worst case location (which is the radial center on the bottom) the Pu-239 MDCs for the paper and latex matrices exceed that standard for a 1,000 sec count. An extended count time would be one method to produce results that would meet the standard. However, another approach for declaring plutonium contamination is below a certain threshold is to estimate its activity based upon the known Pu-239 to Am-241 activity ratio. This ratio is frequently well established in aged weapon’s grade plutonium samples and often falls in the range of 7:1 to 10:1. Therefore, if the detection limit for Am-241 is at least a factor of ten below the standard (i.e., <0.011 Bq/g or 0.3 pCi/g), then one can confidently declare that the Pu-239 contaminants are also below the 0.11 Bq/g (3.0 pCi/g) standard. Detection limits for Am-241 are always well below 0.011 Bq/g (0.3 pCi/g) for all matrices in a 1,000 sec count time.

It is also interesting to note that the Pu-239 detection limit for 4 kg of shredded paper is 71% higher than the detection limit for the same mass of tyvek coveralls. Furthermore, the Pu-239 detection limit for 8 kg of latex gloves is only 17% higher than the detection limit for one-half the mass of paper. On the other hand, the measured detection limits for Am-241 in the same three matrices vary from one another by less than 4% (10.7 Bq to 11.1 Bq).

The conclusions to be drawn from these comparisons are somewhat obvious. The attenuation affects on a 59.5 keV gamma ray in low-density material are not overly dependent on the specific chemical makeup of the material as long as it consists of low-Z materials. However, the specific chemical composition of low-Z material can have a very large impact on attenuation losses for x-rays in the 13-21 keV region. Hydrocarbons like polyethylene consist largely of hydrogen and carbon atoms (Z = 1 and Z = 6, respectively). Meanwhile, in addition to hydrogen and carbon, latex also includes nitrogen atoms (Z = 7), and paper includes oxygen atoms (Z = 8). At 59.5 keV there is very little difference between the attenuation properties of hydrogen, carbon, nitrogen, and oxygen – however, as Fig. 2 exhibits, there are considerable differences in the very low-energy region from 13-21 keV.

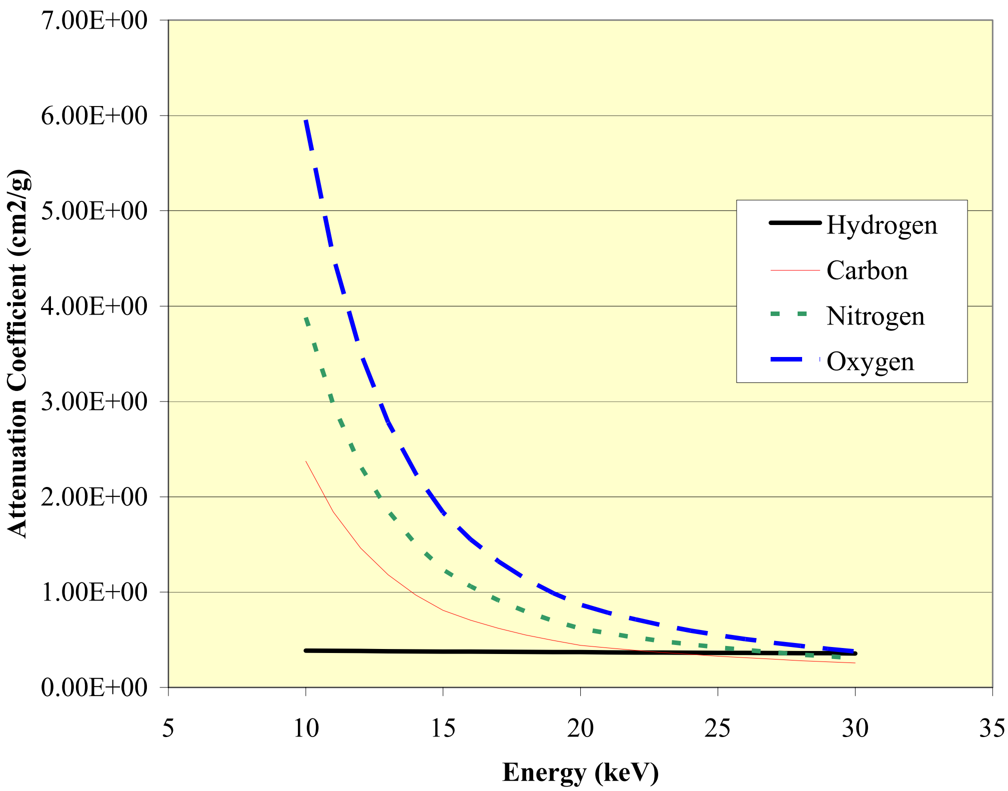

Fig. 2 Mass attenuation coefficients at very low energies

During the course of this work, we designed a simple method to compare the low energy attenuation losses of materials not included in these tests to the materials that have already been evaluated. We carefully assembled a rectangular block of each distinct material with an identical area density (i.e., 0.82 g/cm^2). Next we placed an Am-241 source on the far wall of the ZEUS detection chamber to determine the unattenuated count rates for the low-energy x-ray and 59.5 keV gamma ray ROIs in our detector.

Then we collected another spectrum with each individual block of material covering the Am-241 source to determine the attenuation losses in the x-ray and 59.5 keV ROIs. Since each block of material has the same area density the differences in attenuation losses are caused by differences in mass attenuation properties of each material. This simple method allows us to avoid the arduous task of measuring each material's uniform calibration factor to determine if detection limits are acceptable for the GIC program. The results of these simple attenuation measurements are presented in Fig. 3 for the following materials: polyethylene, latex, paper, and pylox nylon.

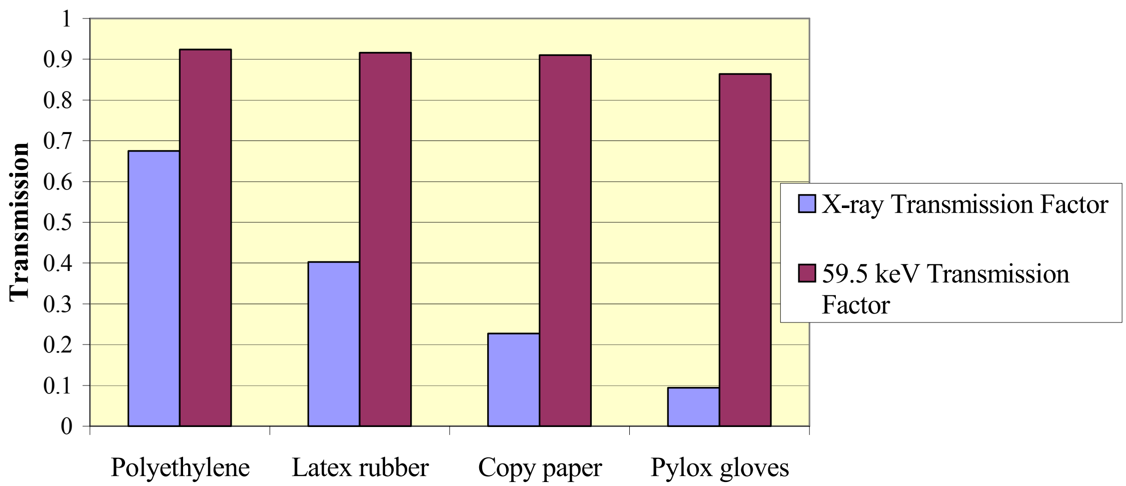

Fig. 3. Photon transmission through 0.82 g/cm^2 of various materials.

As Fig. 3 depicts, our transmission measurements of pylox gloves (a matrix material that was not previously evaluated) revealed that low-energy x-ray attenuation losses are quite a bit worse than they are for other materials that we've evaluated. Based on these results, unless we relied on the 59.5 keV Am-241 gamma ray as described above, we would be hesitant to perform free-release measurements on matrices of pylox gloves with the potential for Pu-239 contamination, especially if we were hoping to achieve detection limits below the suggested screening level of 0.11 Bq/g (3 pCi/g) in ANSI/HPS N13.12. At this writing we are not yet sure what specific atoms are included in the chemical makeup of pylox gloves so we are unable to offer an explanation for the large attenuation losses being caused at low energies.

SUMMARY

The ZEUS system has been demonstrated to be extremely sensitive to the detection of Pu-238, Pu-239, and Am-241 in low-density matrices. By and large these detection limits are below the volume clearance levels proposed in ANSI/HPS N13.12 for a 1,000 sec count time. Furthermore, we have developed a simple method to determine if other materials will be acceptable for inclusion in the GIC program at some future time.

REFERENCES

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