Q^2 - A VERY LOW LEVEL QUANTITATIVE AND QUALITATIVE WASTE ASSAY AND RELEASE CERTIFICATION

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INTRODUCTION

Radioactive waste disposal is an expensive operation for nuclear power plants and other users of large quantities of loose radioactive material. A major volumetric component of this is generated from the practice of establishing radioactive contamination control zones, and assuming that everything in the control zone is contaminated. Since many of these items are of little residual value, it is common practice to dispose of them as radioactive waste without attempting to evaluate whether they are clean. With today's cost of radioactive waste disposal this is an expensive practice.

A typical nuclear power plant generates 300 m^3 (10 000 ft^3) of solid low-level radioactive waste per year. It is estimated from observations of waste generating practices and from a survey of reactor health physics supervisors, that over 50% is uncontaminated. Average waste disposal costs in the United States in 1987 were nearly $500 per 200 L (55-gal) drum. This includes the cost of the container, labor, documentation, transportation, and disposal fees. They are higher today, and will no doubt be higher in the future. If a device could economically and reliably prove that only 25% of the waste were clean, then the potential cost savings would be over $175,000 per year.

REGULATORY ENVIRONMENT

Current United States' regulations do not have a quantifiable limit of radioactivity to define "clean" or "uncontaminated". However, 10CFR20.302 allows (and even encourages) specific license amendments to exempt certain waste streams from radiological controls. It is up to the applicant for such exemptions to propose and defend the maximum concentration of radionuclides in the waste stream.

In order to estimate the magnitude of potentially acceptable release limits, consider the following derived comparisons. Table 1 lists the maximum level of ^{137}Cs and ^{60}Co in a 20 kg bag of waste, if compliance is based on the indicated reference condition.

Table 1.
Reference Condition and Derived Release Limit

REFERENCE		LIMIT IN 20 kg SAMPLE	
US NRC Reg Guide 1.86; 15 000 dpm/100 cm; all activity in sample in 100 cm^2 area		8 nCi	(300 Bq)
	^{60}Co	6 nCi	(220 Bq)
10CFR20 MPC-water Non-occupational limits	^{137}Cs	4 nCi	(150 Bq)
5 mrem/year, assuming waste is uniformly distributed as soil and used for gardening and home construction	^{60}Co	6 nCi	(220 Bq)
	^{137}Cs	20 nCi	(750 Bq)

Admittedly, these are quite conservative, and it is possible that release limits an order of magnitude or so higher could be supported by more detailed analyses. However, even with higher maximum limits, measurement capabilities at 10% of the release limit are a normal practice to assure compliance. It is concluded, therefore, that a reasonable sensitivity goal would be 400 Bq (10 nCi) per sample.

SYNOPSIS OF PROPOSED SOLUTION

There are two key elements in an effective waste volume minimization program. The first is segregation of potentially clean waste at the source of its generation. An effective waste segregation program will include training of personnel and multiple color coded receptacles in the waste generation area. This allows those most knowledgeable of the expected level of contamination (the generators) to place materials likely to be clean in one container, and materials known to be unreleasable in a different type of container. Perhaps an intermediate level container could also be used. Segregation at the source reduces cross-contamination of clean material. A successful waste segregation program then allows the measurements to concentrate on the expected clean and releasable waste.

The second element is a direct measurement program. The device should be economical to operate and use a minimum of labor. Beta counting is labor intensive requiring bags to be opened, separating the waste and, therefore, costly. One way to save labor is to use gamma counting and count the waste containers (typically plastic bags) the way they are received.

A gross gamma (non-spectroscopic) counting system would be less expensive, however, spectroscopy has enough advantages to warrant the extra cost. At the low measurement levels speculated in Table 1, gross non-spectroscopic counters will prove to be less effective in releasing material as clean, as this is the same level of natural contaminants in common materials. A typical 20 kg soil sample, perhaps representative of floor sweepings, will contain 800 Bq (20 nCi) each of ^{40}K, ^{226}Ra, and ^{232}Th. Paper, vegetation, and wood all have ^{40}K at a nominal 1500 Bq (40 nCi) per 20 kg sample. Gross counting would unnecessarily reject this natural radioactivity. Properly designed spectroscopic counters, however, will not include this natural radioactivity.

In addition to rejecting fewer bags as contaminated, the output of gamma spectroscopic analysis can be used to properly characterize the waste. One of the problems with this type of radioactive waste is that it is not homogeneous. Waste generated from one section of the plant, or during one period of operation, will likely have a different mix of radionuclides than waste from other sections or times. Radioactive waste is required to have all major radionuclides and several specific radionuclides quantified to comply with 10CFR61. Since all nuclear power plants in the United States conduct extensive radionuclide characterization of major waste streams, the ratios of these various nuclides are reasonably well known and can be used to establish scaling factors.

Gross counters (beta or gamma) must assume a particular mix of radionuclides. However, if gamma spectroscopic results are available, more accurate scaling factors can be used that are a function of the measured radionuclides. ^{90}Sr is more precisely correlated to ^{137}Cs activity than to gross counts from whatever nuclides are present. As a result, the total accuracy of measured and inferred nuclides is better for spectroscopic systems.

Gross beta counters will miss or underreport radioactivity that is covered by any absorber, and will have extremely low sensitivity to the remaining gammas. Gross gamma counting will likely miss low energy nuclides, as they will be near the noise threshold, especially in elevated backgrounds. Since these units will probably operate in elevated background areas, this can become a serious problem if the radionuclide mix is not stable.

REQUIREMENTS FOR A GAMMA SPECTROSCOPIC RELEASE CERTIFICATION SYSTEM

There is no legal requirement for such a sophisticated device as the Q^2. However, a properly designed system will accomplish two important things: It will save the licensee money over other measurement devices, and it will generate higher quality results.

To maximize monetary savings, the device must be very easy to operate by inexperienced semi-skilled labor. It must be spectroscopic so natural radioactivity will not increase the rejection rate. The system must have adequate sensitivity for short counting times (10

minutes or less). To avoid additional labor, the counter must be able to operate where the low level waste is normally stored. This will be in an elevated background so good shielding all around the sample and detector is necessary.

High quality results are extremely important in today's regulatory and political climate. Since the purpose of this device is to allow the release of known radioactive materials (albeit very low levels), it is very important that the results can be defended as correct. This requires an automated data analysis that is protected from unauthorized modifications, and that stores an auditable computer record of all parameters and data. It must have an easy to use and interpret Quality Control program and internal error checking. The analytical results must be accurate. Since the distribution will be non-uniform, the efficiency should not vary with different source locations within the sample. Finally, the technical algorithms used and the system performance must be well documented and the maximum sources of error must be characterized.

DESCRIPTION OF THE Q² SYSTEM

As shown in the sketch in Figure 1 and the photograph in Figure 2 the shield completely surrounds the detector and sample in all directions. It is constructed of Canberra's low background steel that is free of ^{60}Co. Standard shield thickness is 10 cm (4 in.) but the cavity can hold 15 cm (6 in.) of steel or lead. This allows extra shielding to be used on one or more sides for special situations where there are high activity sources in that direction. The interior of the shield is completely covered by a stainless steel liner. All seams are welded to make it water-tight and allow complete decontamination.

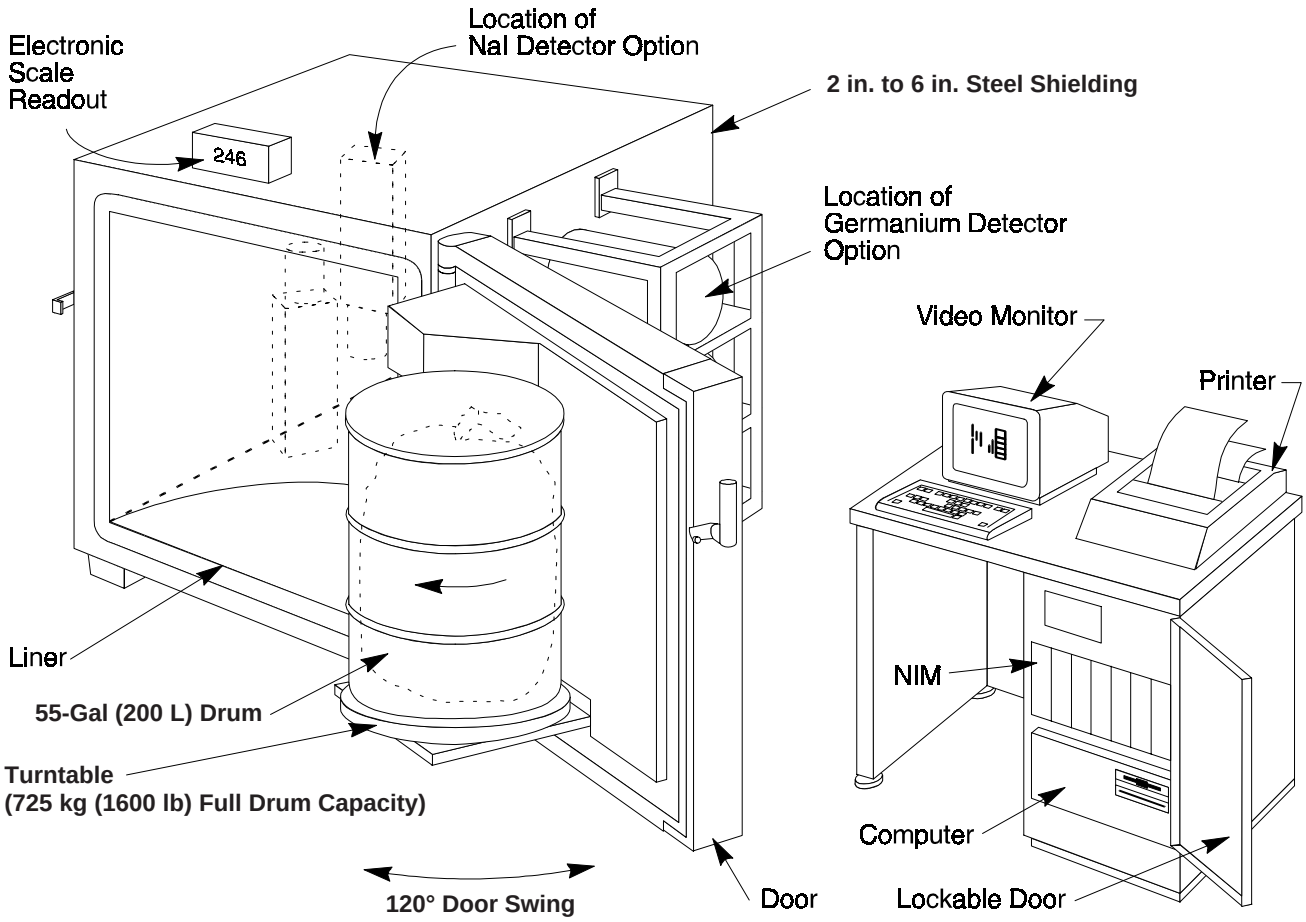

Figure 1.
Sketch of Q² System

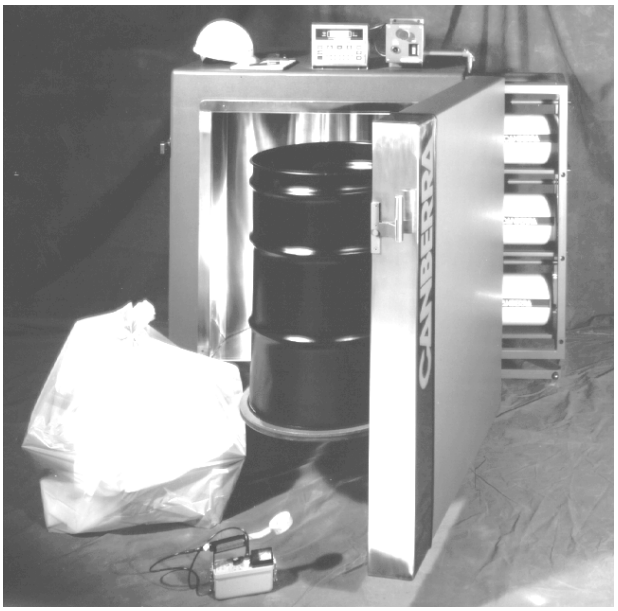

Figure 2.
Shield and Detectors

The door is hinged and is very easy to operate manually. There is a safety device to prevent damage to fingers by the closing door.

Mounted on the door is a rotating turntable. The sample is placed on the turntable which rotates at a nominal 10 RPM. The sample capacity can be a full drum of up to 3.5 g/cc density, as the turntable design capacity is 725 kg (1600 lb). The expected typical use will have an empty drum mounted as a sample container, and bags of waste will be loaded manually.

An electronic load cell mounted on the turntable weighs the sample, and displays the result. The weight and an operator's estimate of how full the drum is, are used to compute density. The density is used to determine the proper efficiency calibration to use.

The detectors are three intrinsic germanium detectors with 7.5 liter multi-altitude cryostats. These are mounted with the detectors inside the shield nearest the sample for maximum sensitivity. The Dewars are outside for convenient filling. Typical detector size is 20% relative efficiency, but larger (better efficiency) or smaller (lower cost) detectors can be used.

The system was also designed to hold up to four each 7.6 x 12.7 x 40.6 cm (3 x 5 x 16 in.) NaI detectors. These would be for extremely low level analyses and shorter count times. The NaI detectors will be thermally isolated for gain stability, and could be thermally stabilized for more severe temperature environments.

The data acquisition system contains a PC-based MCA system, appropriate high voltage power supplies and amplifiers, and operates on an IBM PS/2 30-286 computer. The spectrum from each detector is stored separately. The electronics will be rack mounted behind a lockable door to prevent unauthorized changes. The operator will have access to the keyboard, video monitor and printer, but must use a key to use any other electronic device.

The Q^2 software is an adaptation of Canberra's well documented field proven ABACOS-II software. This contains all of the essential elements of a technically sound, legally defendable low level gamma spectroscopy program. The library driven peak search and Gaussian fitting routines have been shown to be essential for the reliable detection of near LLD peaks.

Routine operation of the software is extremely simple. The computer automatically boots to the Q^2 program. The operator enters his/her name and counter number. The counter number is an identifier for a predefined combination of detectors, count times, calibration, libraries, report styles, etc. Multiple counters can be defined for different predefined situations. The operator then selects one of four different (predefined) sample count times or one of four different QA counting protocols. After selection, the MCA parameters are automatically set up, and the count starts. During the count time, the operator answers a few basic questions about the sample (identification, weight, % of container occupied by sample, other comments desired). Everything else is automatic. When the count is done, the spectra and all analysis parameters are stored on disk. The spectra are analyzed using predefined parameters. The efficiency is assigned based upon a computed density. A full quantitative gamma spectroscopic analysis is performed and the results written to disk. While this report could optionally be printed out, it is expected that most users will select the simple good/bad report for the operator display and print out a simple gummed label to put on the sample.

The analytical results will classify each sample into one of three groups (good, questionable, bad). The system manager defines the maximum allowable quantity of each nuclide. The analysis routine computes the ratios of each nuclide's result to its maximum allowable, and sums these ratios. This sum is tested against a "good" parameter (e.g. less than 0.5) and the "bad" parameter (e.g. greater than 0.8). Samples where the sum is between the limits (e.g. > 0.5 but < 0.8) will be reported as questionable.

In addition to reporting the value of measured nuclides, the software can also derive the activity of other non-measured nuclides which are expected to be present. Typically, each facility has conducted radiochemical sample analysis of the waste. Analysis of these results will determine the best nuclide correlators and the proper formula for correlation. For example, it might be shown that ^{90}Sr (which emits no gammas) is correlated to ^{137}Cs and is typically at 10% of the ^{137}Cs activity. ^{90}Sr can then be added to the library as a derived nuclide, correlated to ^{137}Cs, a 10% factor. Each report with ^{137}Cs activity. If ^{137}Cs is not found, the LLD of ^{137}Cs and the derived LLD of ^{90}Sr will be reported.

Although the software is normally operated in the predefined batch operations mode, the individual steps can be manually performed. Under manual operation, the operator may select certain parameters directly (e.g. count time, key analysis parameters, library to use, etc). All sensitive functions are password protected however, to protect the integrity of the data. Each of the password protected areas can have a unique password, so they can be selectively distributed.

All data are stored on the disk for future use or reanalysis. In addition to the complete data set which is keyed to the automatically assigned file numbers, important portions of the results are entered into an electronic log book. Though the log book can store many thousands of results, the data will periodically have to be transferred to archive media to clear the disk. Predefined reports will print out summaries (e.g. all samples of type A counted in January) and total the activity of each nuclide.

The software also contains a fully integrated Quality Control program. Periodically the operator counts a check source in a reproducible geometry. The program tests the results against predefined acceptable limits of gain, peak shape, and activity. The operator is immediately notified of results that are out of range, and all data is entered into the QC historical file. The Quality Control program also monitors and records background check results and duplicate count results.

Q² SYSTEM PERFORMANCE

Four calibration drums were prepared, each a different density (0.1, 0.3, 0.8, and 1.7 g/cc). This is representative of uncompacted waste (0.1–0.3 g/cc), compacted waste (near 0.8 g/cc) and solidified wastes or soil (1.5–3.0 g/cc). A multi energy source (^{152}Eu) was used to generate different efficiency calibrations for a uniform distribution. Figure 3 shows a typical set.

From knowledge of the system efficiency and background at each energy it is possible to compute a generic Lower Limit of Detection function. The LLD was computed at each energy for and empty shield background using the formula in Equation 1.

$$LLD = \frac{2.71 + 4.66 \cdot B^{1/2}}{E \cdot T \cdot F} \qquad \text{(Eq 1.)}$$

Where:

LLD = Activity at which there is a 5% chance of false positives and a 5% chance of false negatives.

B = Number of counts in the background spectrum where a peak would be if it were present.

E = Efficiency.

T = Sample count time.

F = Conversion factor for desired activity units.

Note that there is no correction for gamma yield at this point. Therefore, the LLD assumes 100% yield.

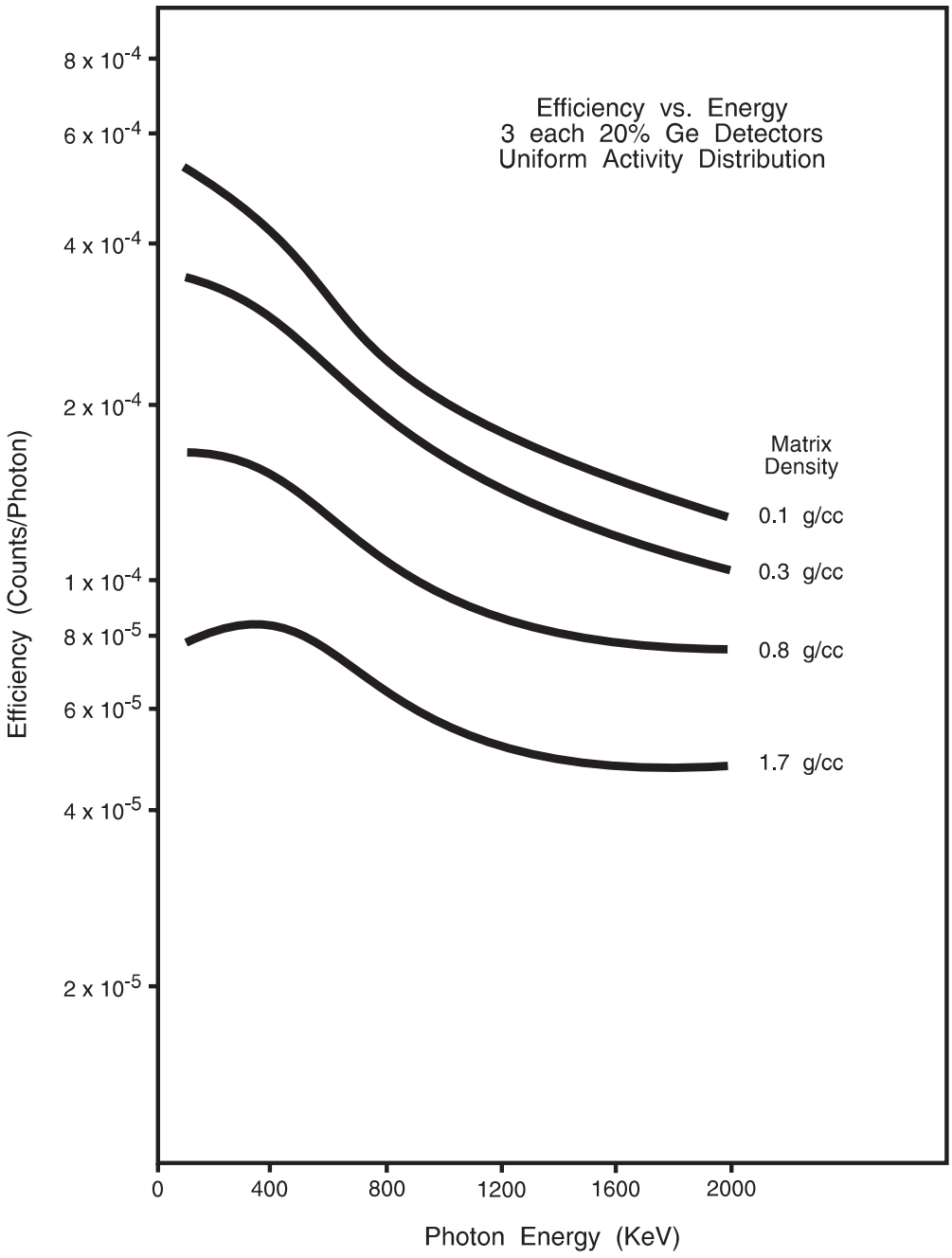

Figure 3.
Efficiency vs. Energy

Figure 4 shows a typical LLD as a function of energy for four different densities. This is for a 10 minute count time. These curves assume 100% gamma yield, therefore for any specific nuclide, the graph must be evaluated at the appropriate energy and the result divided by the fractional gamma yield for that energy. Shorter count times than 10 minutes can be used, but that will raise the LLD somewhat. However, in general 200 Bq (5 nCi) per sample of most nuclides can be detected in a five minute count of low density waste. On concentration basis, this represents 4 Bq/kg (108 pCi/kg) for ^{137}Cs. For high density materials, the activity LLD increases to 550 Bq per sample (15 nCi) but the concentration LLD decreases to 2 Bq/kg (54 pCi/kg).

Accuracy of results is an extremely important consideration. The samples are not expected to have a uniform radionuclide distribution. It is possible (likely) that most of the activity is on a small item in the larger sample. For release certification, it is essential to know the worst case uncorrected error. To determine this, a multi-energy point source was moved throughout each of the inert calibration drums.

As the source is moved vertically, the efficiency changes somewhat with position. It is maximum in front of each detector, and at a minimum between the detectors. The distance between the detectors has been optimized to minimum variations of efficiency vs. source height. This is typically ± 20%.

A much more important source of error is source location vs. radial distance from the drum surface. A source at the outer surface of the drum will have higher efficiency than the source at the axis of rotation. These variations were evaluated as a function of source location, matrix density, and energy. Figure 5 is a family of curves that defines the maximum efficiency error from non-uniform distribution. This assumes that 100% of the activity is contained in a 1 cm diameter sphere placed at the worst location. For low density samples this maximum error is quite reasonable (a factor 1.3). For high densities, especially at low energies, the maximum error increases significantly.

Fortunately, most medium and high density wastes are likely to be adequately homogeneous. Compacted wastes are typically shredded first which mixes them. Solidified wastes have been mixed, and soil samples also are likely to be homogeneous. The maximum errors of Figure 5 must be evaluated to see of they apply, and then appropriately factored into the analysis error calculation and release limit parameters.

CONCLUSION AND PLANS FOR FURTHER IMPROVEMENTS

It has been shown that a gamma spectroscopy system can be developed that is extremely easy to use, and very sensitive. Samples in the 200–400 Bq (5–10 nCi) range can be detected with a 5–10 minute count time. For low and medium densities, the maximum error is less than a factor of two, no matter where the sample is placed in the counting volume.

Full analysis has not been completed with NaI(Tl) detectors, but they are expected to have the same Lower Limit of Detection, but with a one minute counting time. Accuracy is expected to be the same as for the germanium system.

Techniques are available to reduce the maximum uncorrected error. Increasing the detector-sample distance and collimating the detector field of view will both help. Unfortunately, both of these lower efficiency and therefore raise the LLD. A third technique is under investigation that uses the variation in count rate as the sample rotates to compute a correction factor for non-homogeneous distribution. However, for use in assay of intermediate and low density materials, these techniques seem unnecessary.

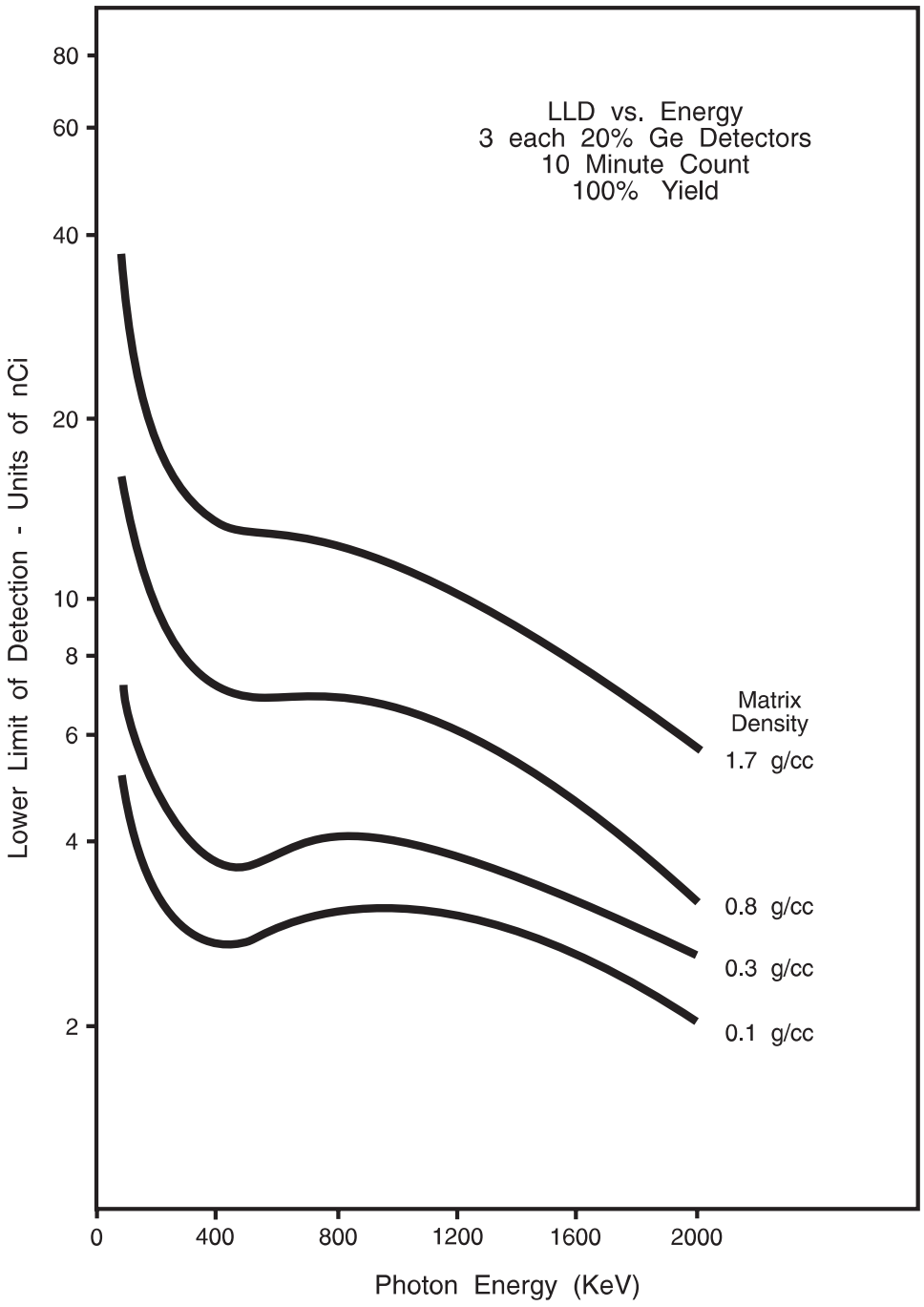

Figure 4.
Lower Limit of Detection

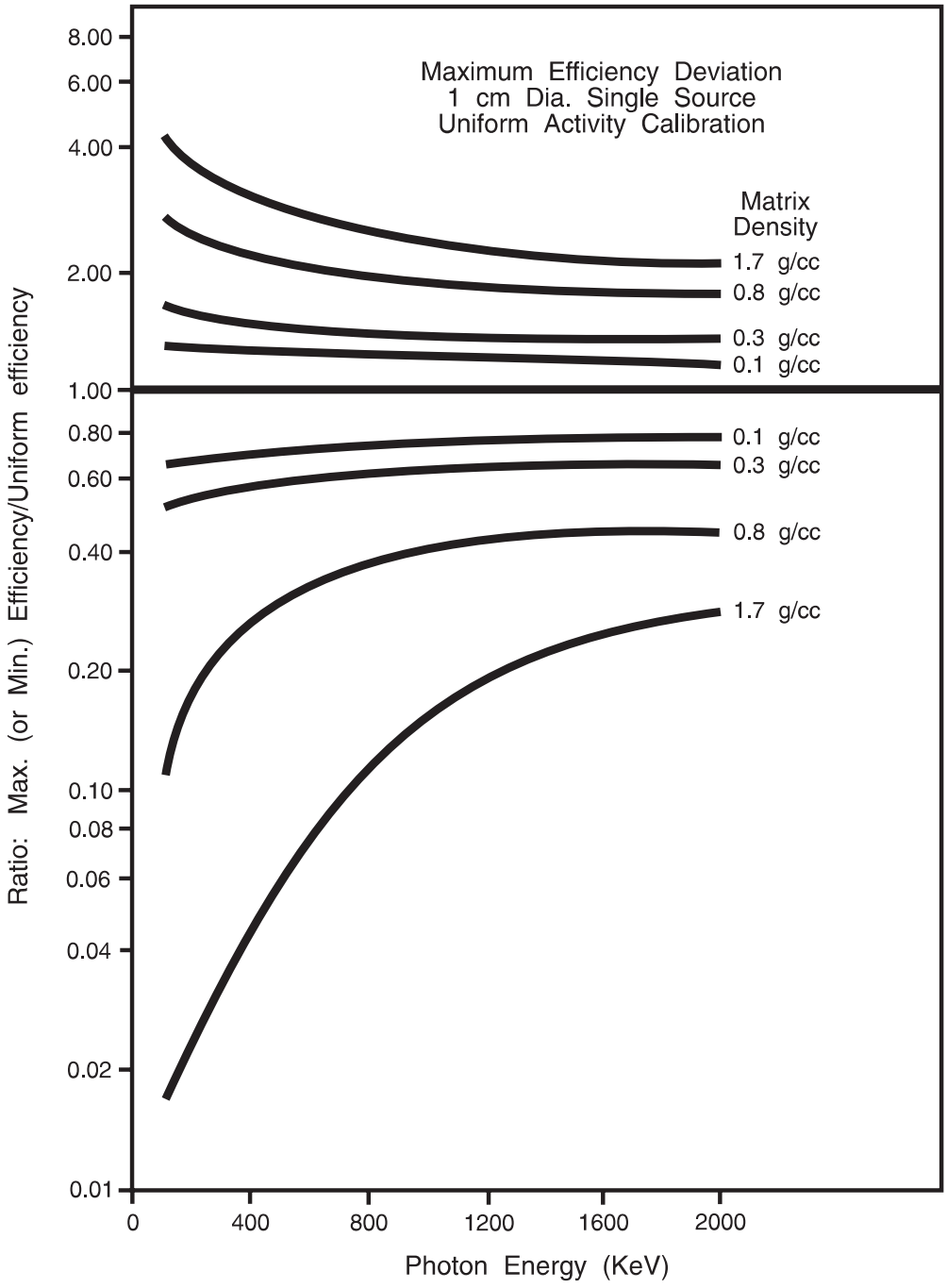

Figure 5.
Maximum Efficiency Error

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ABSTRACT

Low level radioactive waste disposal is very expensive, especially when all of the handling, transportation and documentation costs are included. However for most generators, a large fraction of this low level waste is not contaminated at all, or only slightly so. This paper describes the development and performance of a low level counter that is convenient to use, and that can accurately identify and quantify the radioactivity of any gamma emitter that can be placed in a 200 L (55-gal) container.

These measurement results can be used to verify the absence of radioactivity at very low levels (10 nCi (370 Bq)/ sample), and to identify the nuclides and quantities present, while differentiating against natural radioactivity (Radium, Thorium, Potassium). These results can be used as part of a 10CFR20.302 waste stream exemption program, and thus allow significant savings and a less than one year payback at a typical nuclear power plant.

The Q^2 system is fully shielded to allow its use in the low level radwaste storage area. The detectors are either 3 Intrinsic Germanium detectors or two large NaI detectors. The software is fully automated for simple operation. Correlation factors can be entered to estimate non-gamma emitters from pre-established correlations to other nuclides. Typical Ge detector sensitivities are 8 nCi (300 Bq) LLD for ^{137}Cs at 0.8 g/cc for a 10 minute count time. A NaI detector system can achieve the same LLD in a one minute count.

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