

# Q<sup>2</sup> - A VERY LOW LEVEL QUANTITATIVE AND QUALITATIVE WASTE ASSAY AND RELEASE CERTIFICATION SYSTEM

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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. The 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 thing that can be placed in a 55 gallon (200 liter) container.

These measurement results can be used to verify the absence of radioactivity at a 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 1 year payback at a typical nuclear power plant.

The Q<sup>2</sup> system is fully shielded to allow it's use in the low level radwaste storage area. The detectors are either 3 Intrinsic Germanium detectors or 2 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 Cs-137 at 0.8 g/cc for a 10 minute count time. NaI detector systems can achieve the same LLD in a 1 minute count.

## INTRODUCTION

Radioactive waste disposal is a quite 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 10000 ft<sup>3</sup> (300m<sup>3</sup>) of solid low level radioactive waste. From observations of waste generating practices and from a survey of reactor health physics supervisors, it is estimated that over 50% is uncontaminated. Average waste disposal costs in the US were nearly \$500 per 55 gallon (200 liter) drum. This includes the costs of the container, labor, documentation, transportation, and disposal fees. These costs were from 1987. 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 US regulations do not have a quantifiable limit of radioactivity to define "clean" or "uncontaminated". However, 10CFR 20.302 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 I lists the maximum level of Cs-137 and Co-60 in a 20 kg bag of waste, if compliance is based on the indicated reference condition.

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 10 nCi (400 Bq) per sample.

## SYNOPSIS OF PROPOSED SOLUTION

There are 2 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 un-releasable in a different type of container. Perhaps an intermediate level container could also be used. This 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 measurement program using gamma spectroscopy. The measurement device

TABLE I  
Reference Condition and Derived Release Limit

| REFERENCE  | LIMIT IN 20 Kg SAMPLE |                                   |
|--|-----------------------|-----------------------------------|
| US NRC Reg Guide 1.86; 15000 dpm/100 cm; all activity in sample in 100 cm <sup>2</sup> area                | 8 nCi                 | (300Bq)                           |
| 10CFR20 MPC-water Non-occupational limits  | Co-60<br>Cs-137       | 6 nCi (220 Bq)<br>4 nCi (150 Bq)  |
| 5 mrem/year, assuming waste, is uniformly distributed as soil and used for gardening and home construction | Co-60<br>Cs-137       | 6 nCi (220 Bq)<br>20 nCi (750 Bq) |

should be economic to operate and use a minimum of labor to operate. Opening the bags and separating the waste for beta counting is labor intensive, and therefore more 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 I, 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 20 nCi (800 Bq) each of K-40, Ra-226, and Th-232. Paper, vegetation, and wood all have K-40 at a nominal 40 nCi (1500 Bq) per 20 kg sample. Gross counting would un-necessarily reject this natural radio-activity. Spectroscopic counters, however, when properly designed 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 US nuclear power plants 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 spec-

troscopic results are available, more accurate scaling factors can be used that are a function of the measured radionuclides. Sr-90 is more precisely correlated to Cs-137 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 an 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<sup>2</sup>. 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 in-experienced 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 (5 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 (albiet 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. The maximum sources of error must be characterized.

### DESCRIPTION OF THE Q<sup>2</sup> SYSTEM

As shown in the sketch in Fig. 1 and the photograph of Fig. 2 the shield completely surrounds the detector and sample in all directions. It is constructed of Canberra's specially manufactured steel that is free from Co-60. Standard shield thickness is 4" (10cm), but the cavity can hold 6" (15cm) 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.

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 1600 lbs (700 kg). 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 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 4 each 3" X 5" X 16" NaI detectors. These would be for extremely low level analyses and shorter count times. The NaI detec-

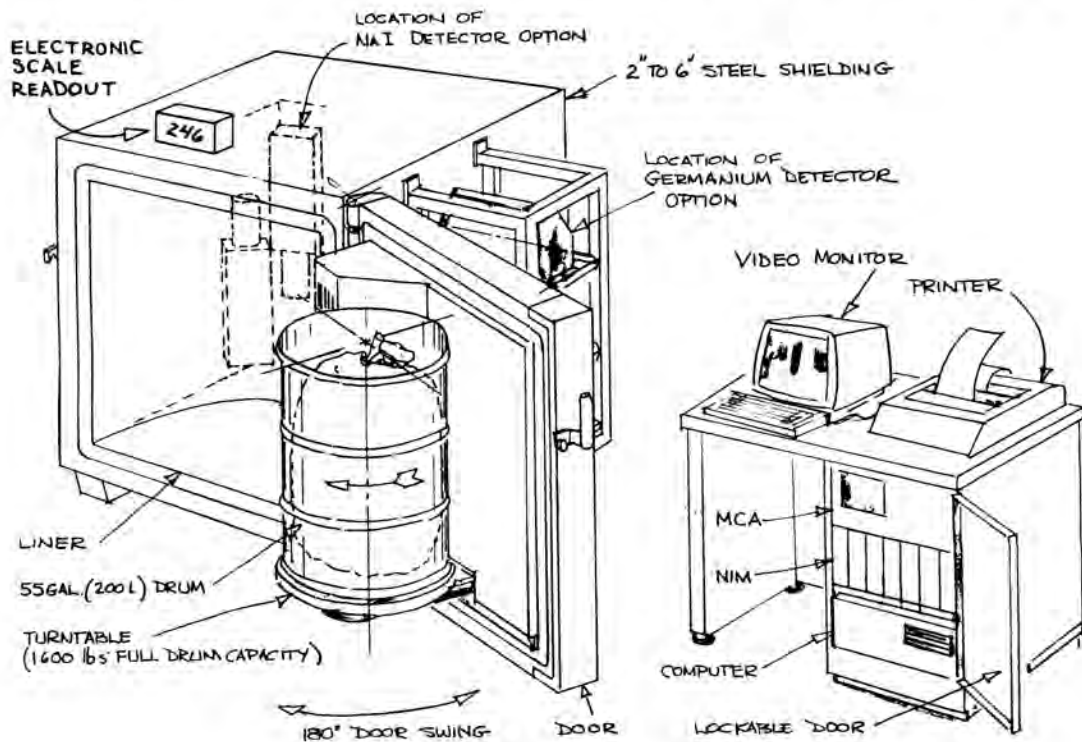


Fig. 1. Sketch of Q<sup>2</sup> System.



Fig. 2. Q<sup>2</sup> Shield and Detectors.

tors 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 PS2 30-286 computer. The spectra 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 utilize any other electronic device.

The Q<sup>2</sup> software is an adaptation of Canberra's well documented field proven ABACOS-II software. This contains all the essential elements to a technically sound, legally defensible 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<sup>2</sup> program. The operator enters his/her name and counter number. The counter number is an identifier for a pre-defined combination of detectors, count times, calibration, libraries, report styles, etc. Multiple counters can be defined for different pre-defined situations. The operator then selects

one of 4 different (pre-defined) sample count times or one of 4 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 is analyzed using pre-defined parameters. The efficiency is assigned based upon 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 3 groups (good, questionable, bad). The system manager defines the maximum allowable quantity of each nuclide. The analysis routine computes the ratios of 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 Sr-90 (which emits no gammas) is correlated to Cs-137 and is typically at 10% of the Cs-137 activity. Sr-90 can then be added to the library as a derived nuclide, correlated to Cs-137, at a 10% factor. Each report with Cs-137 found will also show Sr-90 at 10% the Cs-137 activity. If Cs-137 is not found, the LLD of Cs-137 and the derived LLD of Sr-90 will be reported.

Although the software is normally operated in the pre-defined 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 re-analysis. 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. Periodically, the disk will need to be cleared of the sample data. However, the log book file is very small and

can store many thousands of results. Pre-defined 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 pre-defined 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.

**Q<sup>2</sup> 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 un-compacted 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 (Eu-152) 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 an empty shield background using the formula in Equation 1

$$LLD = \frac{2.71 = 4.66 \sqrt{B}}{E \cdot T \cdot F} \quad (\text{Eq. 1.})$$

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 4 shows a typical LLD as a function of energy for 4 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, 5 nCi (200 Bq) per sample of most nuclides can be detected in a 5 minute count of low density waste. On concentration basis, this represents 0.14 pCi/g (4 Bq/kg) for Cs-137. For high density materials, the activity

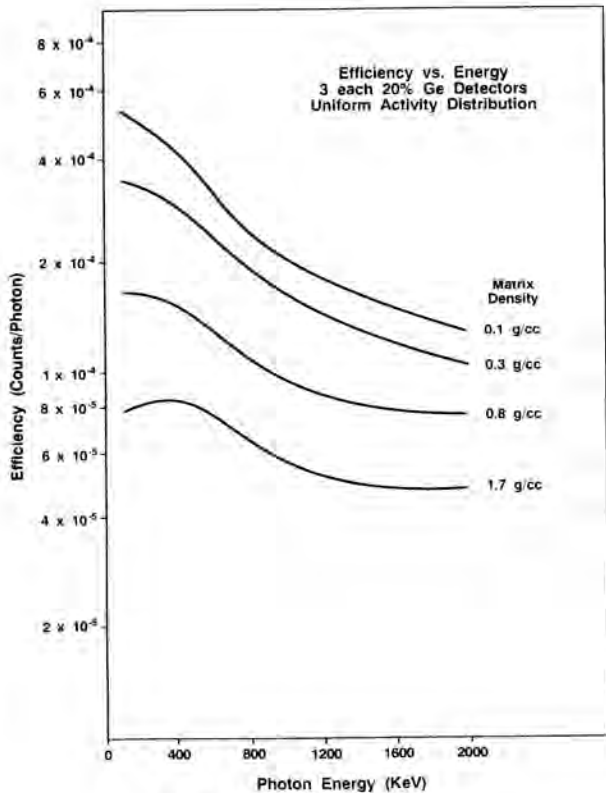


Fig. 3. Efficiency vs Energy.

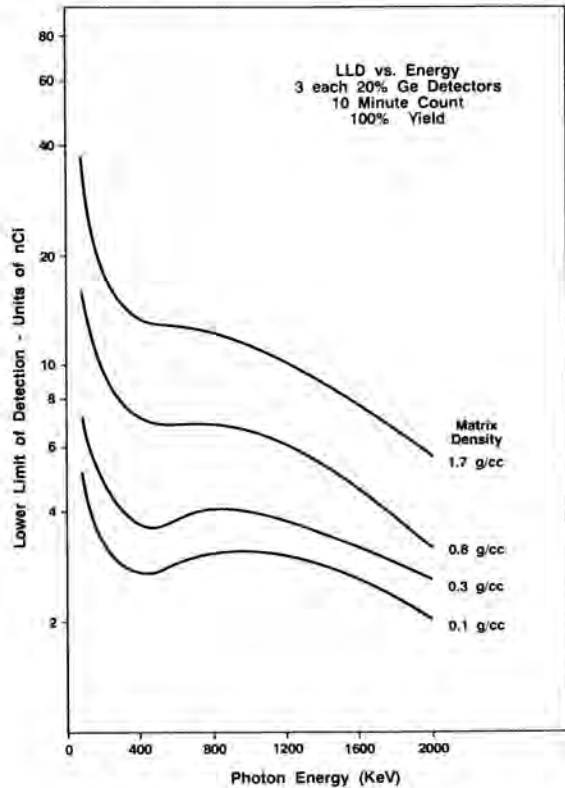


Fig. 4. Lower Limit of Detection.

LLD increases to 25 nCi per sample (550 Bq) but the concentration LLD decreases to 0.5 pCi/g (2 Bq/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 un-corrected 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  $\pm 20\%$ .

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 1cm 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 if 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 5 - 10 nCi (200 - 400 Bq) range can

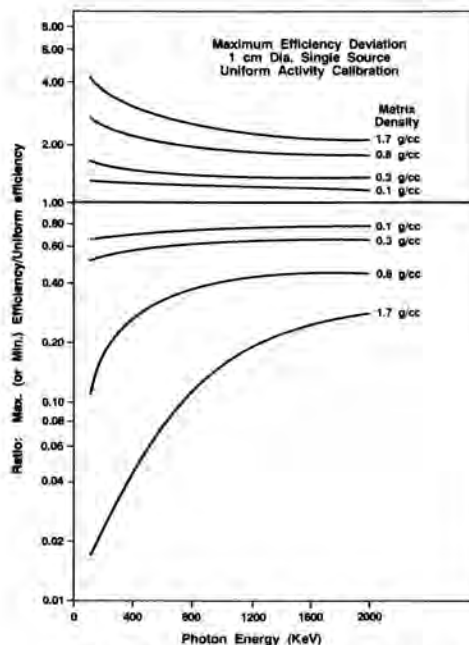


Fig. 5. Maximum Efficiency Error.

be detected with a 5 - 10 minute count time. For low and medium densities, the maximum error is less than a factor of 2, 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 1 minute counting time. Accuracy is expected to be the same as for the Germanium system.

Techniques are available to reduce the maximum un-corrected 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 un-necessary.