Validation of Efficiency Calibration for the Clearance Measurement System - 10086

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ABSTRACT

The "Regulations on clearance levels for radioactive waste management" was promulgated on December 29 2004 by Atomic Energy Council (AEC) of the Executive Yuan. The regulations stipulate the waste release plan that needs to be submitted when reviewing waste released from regulation control; this is based on the waste category and operational history. The primary technique used is based on the non-destructive assay (NDA) of bulk waste and determines the activity concentrations of gamma nuclides within the waste. The Institute of Nuclear Energy Research (INER) set up a release measurement laboratory and a major clearance measurement system. In order to implement the release plan, shielded gamma spectrometry was utilized for waste package counting of metal and concrete debris arising from the decommissioning and dismalting of the Taiwan Research Reactor.

When using a NDA system, a standard source needs to be prepared to allow efficiency calibration and this source should match the geometry and physical property of the sample. However, this is not easy or accurate when calibrating bulk material in large waste drums. Efficiency calibration for waste drums was carried out using the shell-source method and ISOCS mathematical efficiency calibration software; the latter eliminates the need for radioactive sources for efficiency calibration by the use of a Monte-Carlo based mathematical efficiency computing method. Although the efficiency calibration softwere provides a user-friendly input interface in terms of counting geometry, variations and/or errors in the input parameters can result in incorrect calibrations and large deviations from the true value. Therefore, validation of the software used for the efficiency calibration was necessary and this needed to be done in a simple, safe and econmical way. A validation waste drum was packed with KNO_3 powder. This was used as a test medium in this study because this chemical is homogeneous, easily available and contains significant amount of the natural nuclide K-40, which is ecempt from control. Samples from the packing process were collected in an air-tight plastic jar of 125 ml capacity and counted with laboratory gamma spectrometers calibrated using NIST standard reference material. The laboratory results were compared with the counting results obtained for the KNO_3 verification drum by the clearance measurement system using the Grubbs test. The results showed that the ISOCS calibration tool complied with ANSI N42.22 acceptance criteria for traceability to NIST.

Keywords: clearance level, clearance measurement, gamma spectrometer, efficiency calibration, waste

INTRODUCTION

A large amount of waste is produced during the routine operation, maintenance, decommissioning and dismantling of a nuclear facility [1] and this is regarded as radioactive waste because it comes from a controlled area. However, only a very small proportion of this waste will show significant radioactivity based on experience during previous decommissioning projects. Impacts on human health and environmental safety are minimal when handling non-contaminated and very low level contaminated radioactive waste. In this context, if all of waste materials from a site are dealt with as low-level radioactive wastes in terms of management and treatment, the principle of cost control and resource utilization can not be fulfilled. The "below regulatory concern" concept for handling low-level radioactive waste (radwaste) has been receiving increased international interest over the last few years. The RS-G-1.7 safety guide, "Application of concepts of exclusion, exemption and clearance" was formally completed in August 2004 [2]. The "Regulations on clearance levels for radioactive waste management" was promulgated in Taiwan on December 29 of the same year and follows the IAEA safety guide [3].

Gamma radionuclides, such as Co-60 and Cs-137 with their highly penetrating gamma rays, are contained in most radwaste and can be used when assessing a bulk object by non-destructive assay (NDA) using a gamma spectrometer in order to determine activity concentration. Extracting a truly representative sample for laboratory analysis of the typical radwaste matrix is difficult to do, especially under the common condition where container matrix is not homogeneous. Because of this, NDA of the entire unopened container is one of the common methods used for radwaste assay. Since a large amount of waste is produced during the decommissioning and clearance of a nuclear facility and the release limit is quite low (e.g., 0.1 Bq/g for Co-60 and Cs-137 as designated by the Taiwan IAEA), the shielded gamma counting system was adopted as the key tool [4] for 220L waste drums (or a similar size of waste container) in order to determine if the waste qualifies for free release. The qualitative characterization of gamma nuclides and quantitative analysis of the average activity concentration of nuclides in the waste package can be obtained by the use of a gamma spectrometer. This approach allows direct comparison with the release limit and can be used when assessing release. The Clearance Assay System (CAS) at the Institute of Nuclear Energy Research (INER) has been proven capable of measurement of key nuclides at a detection limit lower than 0.1 Bq/g, which is $1 - 10\%$ of the release limit, when a reasonable count time (300-1000 sec) is used.

In addition to the requirement for sensitivity, validation of the accuracy of the system is also an important requirement. This revolves round how to prepare samples with the same geometry as that of the standard, which is one of the key factors that affect the accuracy of the counting results when using a gamma spectrometer. The maximum sample volume used in the laboratory was about 1L of the reference material in a Marinelli beaker. During the preparation of such types of laboratory standard samples, homogeneity is easy to control when performing the efficiency calibration. However, the consumption of a standard source becomes quite difficult when a 220L waste drum is used due to the very large volume.   Furthermore, the uniformity of the medium and the distribution of the source are not as easily controlled during preparation. Therefore, when a gamma counting system is employed for counting a large waste package, there are three common approaches to efficiency calibration [5].

(1) The simulated standard drum: The reference line sources are inserted at the specific locations in a simulated standard drum filled with different materials (shown as Fig. 1). The drum is rotated for assay during the efficiency calibration process [6] [7] [8].

(2) Mathematical correction from point source: A mathematical efficiency correction is performed for the different energies and geometries based on the differences between the point source standard and sample drums. The most commonly used approach for the hypothetical mode is a point detector and the use of the inverse-square law and matrix attenuation corrections [9] [10] [11].

(3) Monte-Carlo spatial detector calibration followed by mathematical sample geometry and attenuation calculation: First, MCNP is used to create a Detector Characterization for each specific detector used. This Characterization allows the Canberra ISOCS software to know the efficiency for any point in space around the detector. Detailed parameters describing the location of the detector with respect to the sample, any collimators surrounding the detector, the sample container, and the sample matrix are entered into the code. The ISOCS software then computes the efficiency for the container at energies requested by the user [12] [13].

Fig. 1. A simulated standard drum used for efficiency calibration with method 1.

The currently used efficiency calibration at the CAS is method (3). The initial reason was because of the difference in wall thickness between the supplier's simulated standard drum and real waste drums at INER. Using ISOCS, the time and cost for a new efficiency calibration was greatly reduced, the radiation exposure of the operators could be lowered, since there was no need to purchase a standard line source and no need for the drum counting process. In addition the use of the ISOCS software allows us to calibrate for many other different container sizes, shapes, sample fill heights, sample densities, sample matrix materials, etc. The supplier has carried out validation testing of the calibration software using a large number different multi-nuclide reference sources which are many different sizes, shapes, and counting geometries [14].   The uncertainty for k=1 was calculated to be 4.5% for high energies (>150 keV) and 9% for low energies.   The method used to validate the ISOCS calibration created for the CAS is described in the next section.

METHODS

Equipment and material

(1) CAS: A commercial Automated Qualitative and Quantitative Low Level Waste Assay System (Canberra®, abbreviated as the AQ2 low-level waste assay system), was used for 220L waste gamma counting designated for clearance or exemption at the INER [15], as shown in Fig. 2. The CAS primarily consisted of mechanical parts, three 32% relative efficiency of HPGe detectors, nuclear instrument modules (NIMs) and computer control software. The related schematic sketch is shown in Fig. 3.

(2) The laboratory gamma spectrometers: Three coaxial HPGe detectors from Canberra Industries were employed in the laboratory (models: GC4020, GC3018 and GC3020) with related NIMs. A standard source, which had traceability to the National Institute of Standards and Technology (NIST) and had the same geometry, was performed for establishing the efficiency calibrations.

(3) ISOCS mathematical efficiency calibration software and Detector Characterization files for each detector in the AQ2.

(4) Calibration drum of the proper size and wall thickness.

(5) Radioactive material for filling calibration drum: Industrial grade KNO_3 (HAIFA chemical corp., Israel) with a purity of higher than 99% due to its chemical stability.

Fig. 2. CAS system at INER. This view shows the drum in the load/unload position and the AQ2 system in the background with the sliding shield door open.
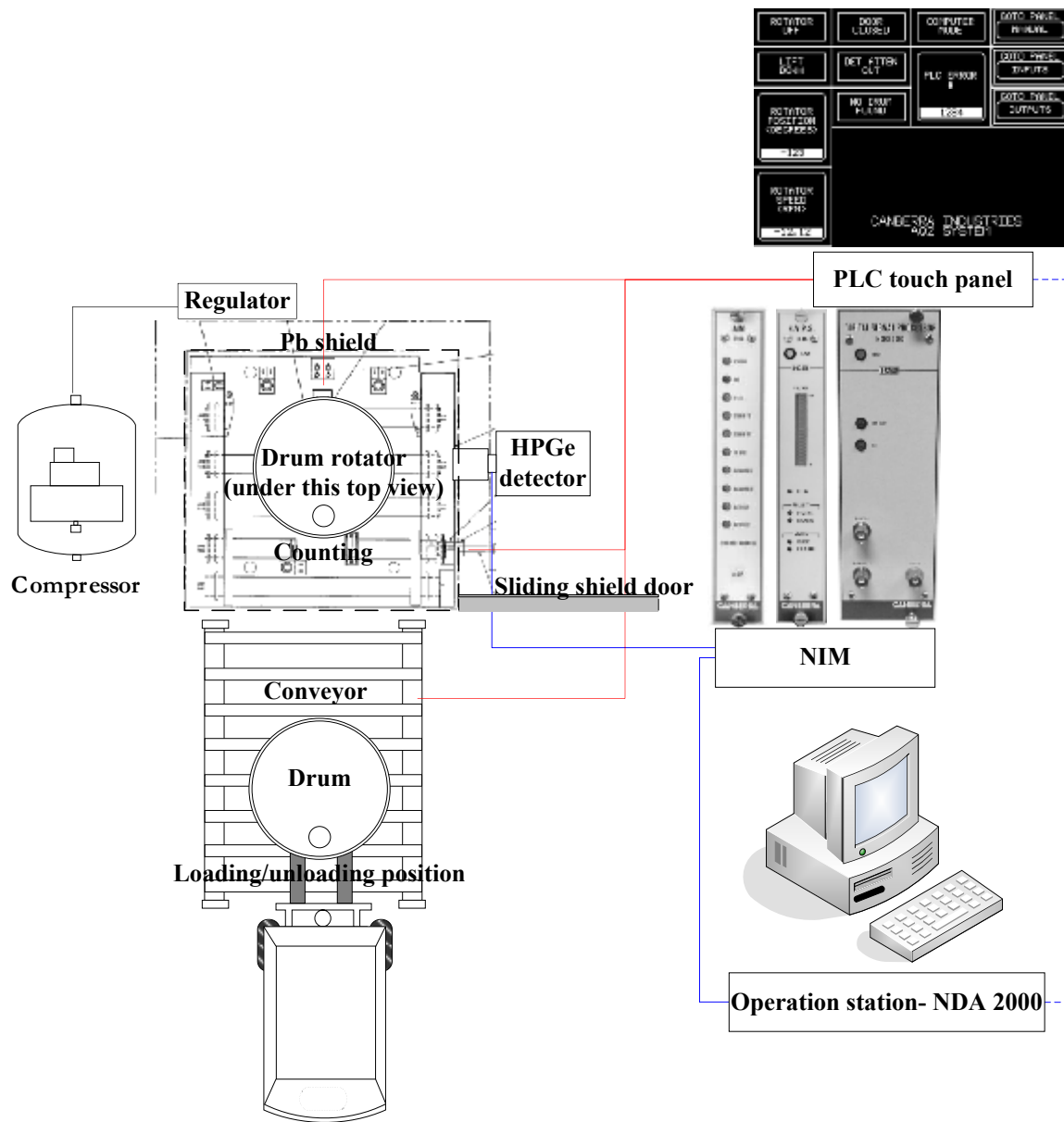
Fig. 3. Major components of CAS.

Validation procedure:

(1) Using the ISOCS software and the simple cylinder template (shown in Fig. 4), the efficiency calibration of the CAS was performed.   Specific parameters of our system were entered, e.g., the diameter, height, wall thickness, and material type of waste drum as well as relative space related to detector. The calibration procedure followed is shown in Fig. 5.
(2) Preparation of the KNO_3 verification drum: Individual bags of KNO_3 were used to fill the drum and each bag was randomly sampled during filling process. Eleven samples were obtained for further laboratory measurement [Fig. 6 (a)].
(3) Homogeneous industrial grade KNO_3 was used to fill the 220L waste drum [Fig. 6 (b)] and the drum was then covered.
(4) The samples from step (2) were measured by the three gamma spectrometers (marked as Lab Det. 1-Lab Det. 3) in the laboratory to provide initial data for analysis.
(5) The activity concentration of K-40 in the 220L KNO_3 verification drum was determined as described in the step (3) using CAS counting.
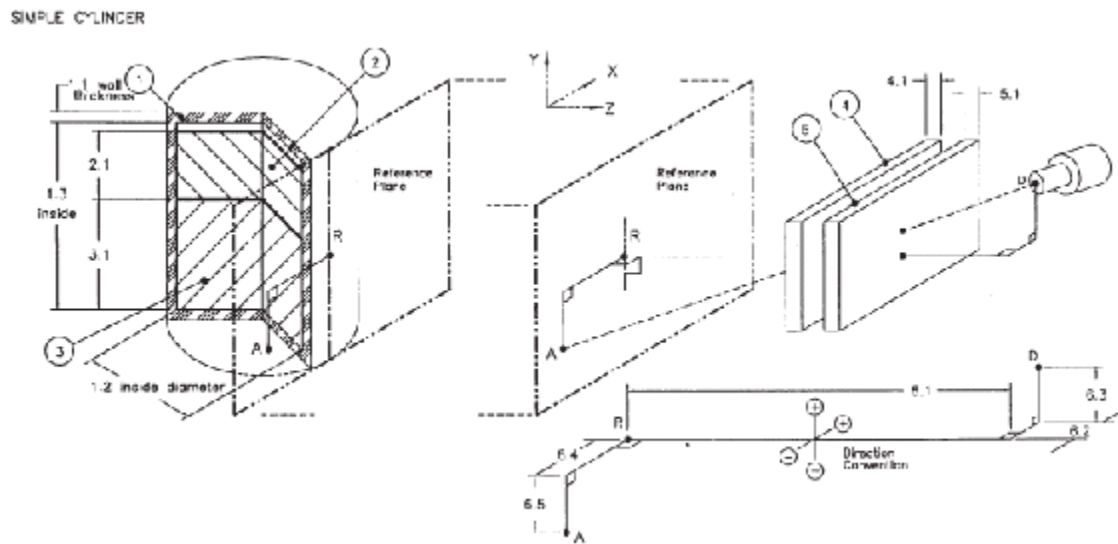(6) The analytical results from two counting systems were compared.

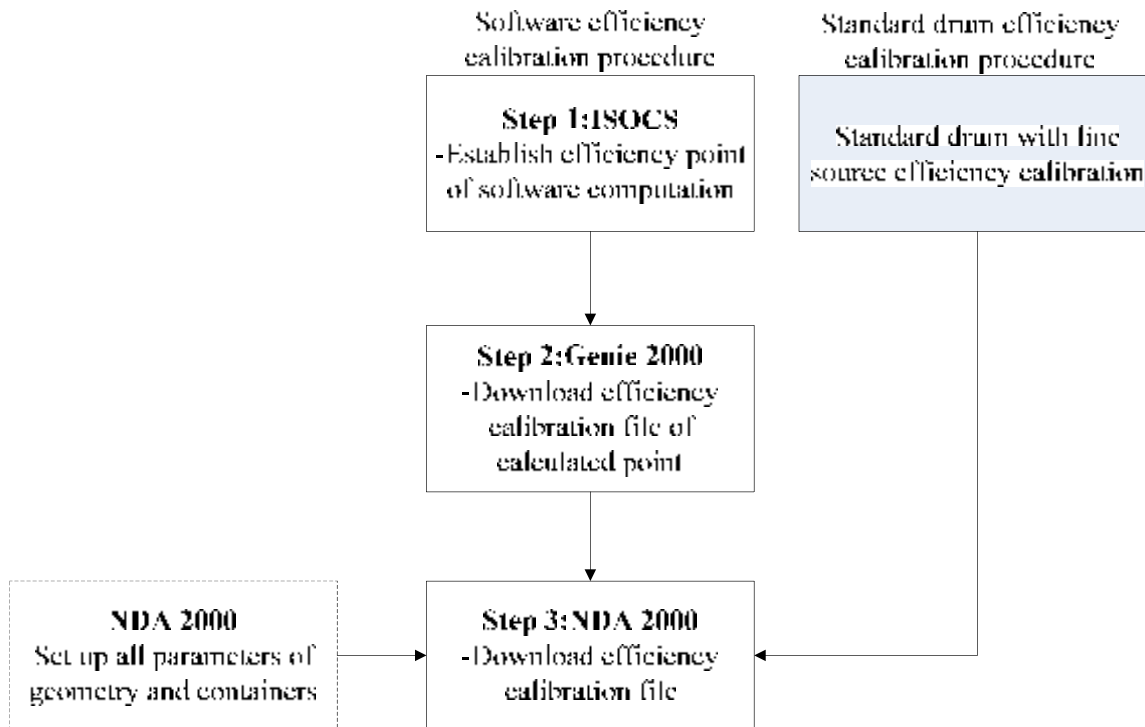Fig. 4. The template parameters of CAS efficiency calibration [8]

Fig. 5. CAS calibration procedure

           (a)   (b)

Fig. 6. KNO_3 subsamples (a) and verification drum (b)

RESULTS

Three different integrated gamma spectrometers were used for the analysis of the 11 samples; these were counted in the laboratory using gamma spectrometry and the system was calibrated using a NIST standard reference. In addition to following the quality control of laboratory procedures, accuracy and NIST traceability are ensured by participating in the annual measurement assurance program of the NEI/NIST MAP and in the CNLA proficiency test. The analytical results and the associated statistical data are shown in Table 1.

Table 1. Sampling analytical results

Sample No.	Results (K-40, Bq/g)		
	Lab Det.1	Lab Det.2	Lab Det.3
1	9.42±0.67 [*1]	9.72±0.73	9.10±0.69
2	9.94±0.70	11.00±0.82	10.10±0.76
3	10.30±0.73	11.30±0.85	10.50±0.80
4	9.97±0.70	10.50±0.79	9.86±0.75
5	9.52±0.67	9.57±0.71	9.31±0.71
6	10.40±0.74	10.60±0.79	10.30±0.78
7	9.81±0.69	10.30±0.77	9.75±0.74
8	9.15±0.65	9.57±0.72	9.61±0.73
9	9.75±0.69	10.80±0.81	10.10±0.77
10	9.90±0.70	10.80±0.81	10.40±0.79
11	9.33±0.66	10.00±0.75	9.36±0.71
Mean value ($\bar{x}$)	9.77	10.38	9.85
Standard deviation (s)	0.39	0.60	0.47
Average relative standard deviation	3.53%	3.74%	3.79%
Standard deviation of the mean ($s_{\bar{n}}$)	0.12	0.18	0.14
Mean value of measurement uncertainty (u)	0.35	0.39	0.37

*1 : Uncertainty based on software data and calculation using the coverage factor, k =2.

Sample homogeneous test: 11 samples were analyzed in the homogeneous test. Grubb's test is generally used for this type of statistical analysis. The calculation is based on a sampling size below 50 and is carried out under the assumption that samples are homogeneous and that a few outliers can be excluded and identified. The statements of Grubb's test [16] are outlined below.

(1) Calculate the mean value of sample (mean, $\bar{x}$) and standard deviation (standard deviation, s).

(2) Calculate the bias of the maximum or minimum in samples (G value), as Eq. 1.

$$G_{max} = \frac{\left| \bar{x} - x_{max} \right|}{s} \qquad or \qquad G_{min} = \frac{\left| \bar{x} - x_{min} \right|}{s} \qquad \text{(Eq. 1)}$$

(3) Judge whether G value is greater than the critical value of the confidence interval (95% or 99%). The outlier should be excluded if G value is greater than the critical value.

(4) Eleven samples were measured by each gamma spectrometer and the critical value of a 95% confidence interval was 2.234, which indicated that uniformity of sample in terms of K-40 distribution is good using Grubb's test.

Analysis of variance for the detectors: If the statistical results by ANOVA analysis or t-test [17] between two detectors according to Table 1 were used to judge whether there was any significant difference between the detectors, the results showed that Det. 1-2 and Det. 2-3 from each other did not pass the test. The uncertainty of the relative standard measurement from a gamma spectrometer was 3.53%-3.79% for a single sample counting, which is higher than

1.2%-1.7% of the standard deviation of the mean ($s_{\bar{n}} = \dfrac{s}{\sqrt{n}}$). Therefore, if the counting

uncertainty is taken into account, the analytical results from three detectors are not significantly different (as shown in Table 2, $t \leq t_{DF,0.05}$). Considering the uncertainty of measurement, the analysis result of the three detectors show no significant difference. Overall, 10.00 ±0.37 Bq/g (k=1) was found based on the mean value and the mean value of the measurement uncertainty from 33 samples.

Table 2. Variance of t-test result for Lab detectors

	Degree of freedom (DF) ^{*2}	t Distribution (95% confidence interval)	t^{*3} (by $s_{\bar{n}}$)	t (by uncertainity measurement)
Lab Det .1-2	18.73	2.10	2.82	1.16
Lab Det. 2-3	20.78	2.09	2.29	0.98
Lab Det. 1-3	21.22	2.08	0.44	0.16
Judgment			Significant difference for Det. 2	No significant difference

$$*2 : \ DF_{1\text{-}2} = \left\{ \frac{\left(\dfrac{s_1^2}{n_1} + \dfrac{s_2^2}{n_2} \right)}{\dfrac{\left(s_1^2 \middle/ n_1 \right)^2}{n_1 + 1} + \dfrac{\left(s_2^2 \middle/ n_2 \right)}{n_2 + 1}} \right\} \qquad \text{(Eq. 2)}$$

$$*3 : \ t_{1\text{-}2} = \frac{\left| \overline{x_1} - \overline{x_2} \right|}{\sqrt{s_{\bar{n},1}^2 + s_{\bar{n},2}^2}} \qquad \text{(Eq. 3)}$$

CAS counting results: The filled KNO_3 verification drum was measured using various count times and the results are listed in Table 3. The mean value is 10.75 ±0.62 Bq/g (n=3, k=1). The reported propagated relative uncertainty did not change with increased count time, because the counting statistics portion of the uncertainty is significantly smaller than the systematic uncertainty used in the calculations. .

Table 3. Results of the KNO_3 verification drum counting by CAS

CAS	Count Time (sec)	K-40 (Bq/g)	Relative uncertainty (k=1)
1	1000	10.75	5.8%
2	30000	10.69	5.8%
3	80000	10.80	5.8%
	Average	10.75	

The results of CAS counting were compared with that of the gamma spectrometers in the laboratory using the ANSI N42.22 acceptance criteria for traceability to NIST for performance testing [18]. This can be expressed by Eq. 4,

$$\left| \overline{x_{CAS}} - \overline{x_{sample}} \right| \leq 3\sqrt{u_{sample}^2 + u_{CAS}^2} \qquad \text{(Eq. 4)}$$

where $\overline{x_{sample}}$ and $\overline{x_{CAS}}$ are the average values from the laboratory gamma spectrometer and

CAS, respectively. The difference in the calculation result, $\left| \overline{x_{CAS}} - \overline{x_{sample}} \right|$ was 0.75 and the

combined uncertainty $3\sqrt{u_{sample}^2 + u_{CAS}^2}$ was 2.17, which meet the acceptance criteria.

DISCUSSION

1. When counting a 220 L waste drum, there are several methods available to identify the activity concentration. The simplest method (AQ2) was used in our CAS. A somewhat more complicated method would be a Segmented Gamma System (SGS), and an even more complicated system would be a Tomographic Gamma System (TGS). The investment cost increases with the increase in complexity of the hardware system, but this increased complexity is sometimes justified as it is expected to to improve the accuracy of the assay, especially for non-homogeneous radioactiivty distributions and non-homogeneous waste matrices. However, since the efficiency is lower, a longer count time is then needed and the drum throughput is reduced, which further increases the cost. Thus, at the present time, there is no need for these more expensive systems. Since a large amount of waste is produced during the decommissioning and clearance of a reactor such as the Taiwan Research Reactor, the appropriate balance in throughput, accuracy and sensitivity must be considered for optimization. The CAS has 3 uncollimated detectors close to the rotating container improve counting efficiency and reduce the counting time. This allows the CAS to be able to handle the large amount of radwaste undergoing clearance measurements. With an average 200 kg in each waste drum, an annual capacity of 540 tons of waste can be passed through the clearance measurement for TRR D&D (decomissioning and decontamination) project.
2. Since a standard drum with homogeneous distribution of a reference radioactive material is not practical to use for the efficiency calibration of bulk waste, the simulated standard drum was used with a traceable line source as shown in Fig. 1. However, error does occur with this type of calibration [6]. Besides the error caused by simulated method itself, errors may come from the differences between the calibration container and the typical containers used at INER. For example the thickness of the standard drum from the supplier was 1.6 mm, whereas the wall thickness of the low-level radwaste drums used at INER was 2.0 mm. Furthermore, the thickness of the container for clearance measurement was 1.0 mm. The efficiency v.s. wall thickness of the waste drum under different densities (d=0.79 - 1.58)

using the efficiency calibration software for Cs-137 is shown in Fig. 7. and a 4%-5.6% error was produced if the simulated standard drum from original supplier was directly used to carry out the efficiency calibrationwithout correction. Much larger errors can happen if the diameter isn't the same for both the calibration and the sample, if the fill height inside the container isn't the same, if the matrix isn't the same.   The use of the ISOCS software allows us to quickly calibrate for each of these conditions to improve the accuracy.

3. In this investigation, the industrial grade of KNO_3, which contains a suitable abundance of the natural radionuclide K-40, was utilized and this reagent was quite succesful when used for the validation of the software efficiency calibration method for CAS. This material has easy availability, minimal regulation, and good homogeneity. In terms of the counting data statistics, direct traceability can be established between the CAS counting results and the national measurement standard even without source preparation for the CAS efficiency calibration. The validation confirmed that the CAS and the gamma spectrometers were in compliance with ANSI N42.22 acceptance criteria for traceability to the NIST (Fig. 8).
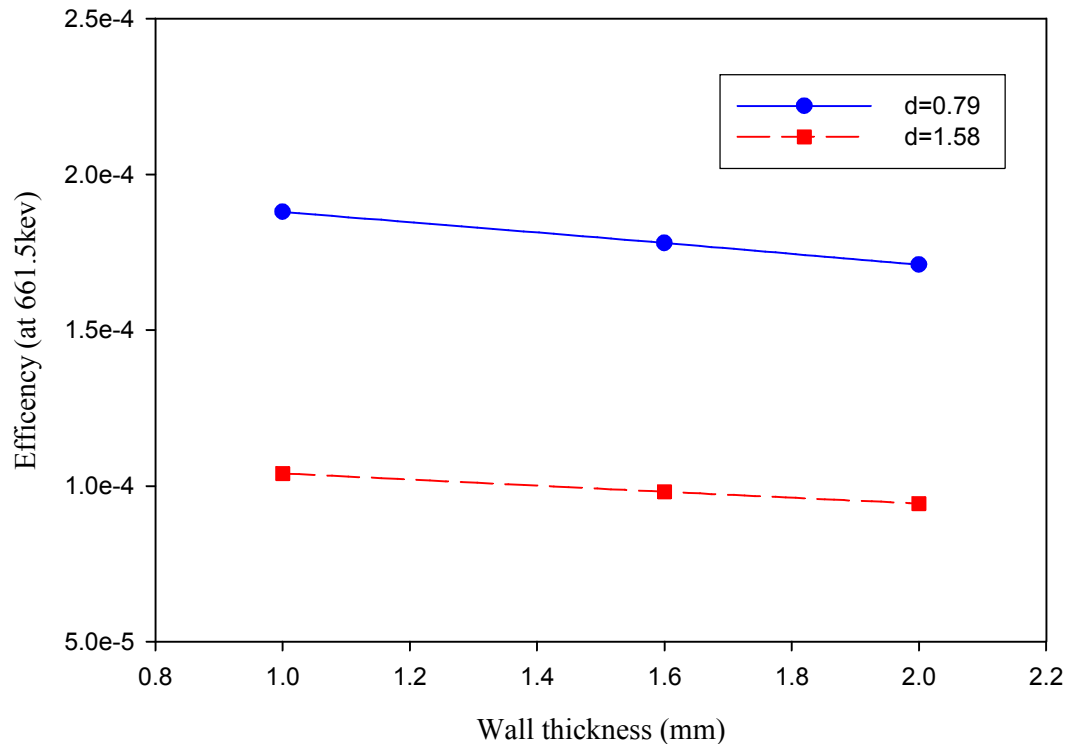
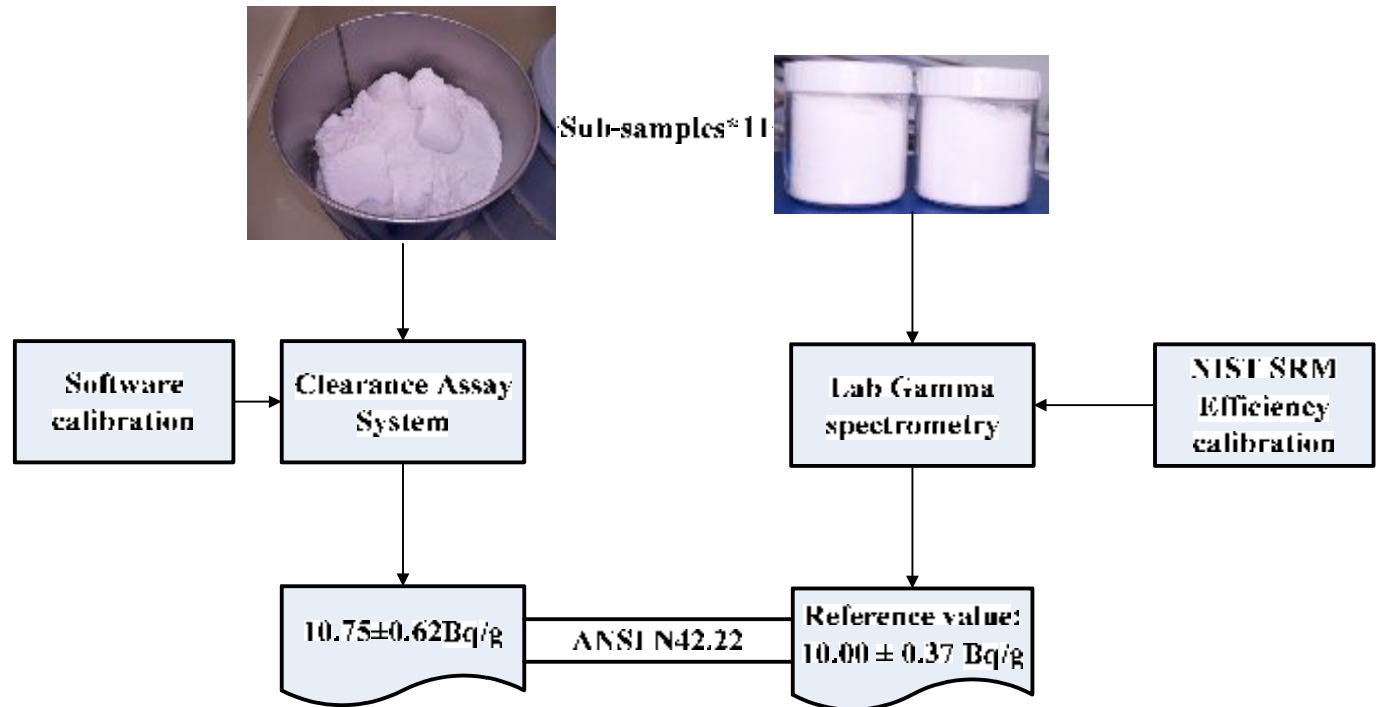Fig. 7. Efficiency curves for the Cs-137 of waste drum with different wall thicknesses

Fig. 8. Schematic diagram of the traceability realtionship between CAS and NIST

CONCLUSIONS

1. The preparation process was very simple and uniform distribution was readily achieved in the drum by the use of a KNO_3 validation drum. The gamma spectrometers in the laboratory were employed for sampling analysis and the counting statistics were used as the reference value in order to verify the measurement traceability for both counting systems. The results showed that the CAS can be traced to NIST. In addition, the accuracy of the CAS measurement and the parameters used in the efficiency calculation give greater confidence to the measured data used in practice for release and also meet the requirements of regulation body.
2. Measurement of activity concentrations of nuclides are quite critical to the decision making when allowing the free release of radwaste. It was important to establish a complete quality assurance system and to ensure the suitability of system in order to enhance the effectiveness of the analytical data. Furthermore, the concept of measurement via multiple approaches can be used to check whether the contamination level of the waste meets the regulations, to reduce the probability of non-conformance and to effectively diminish any impact on the environment and people. The use of the CAS will allow free release and therefore resource re-utilization of the material from the decommissioning of the reactor.

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