PROBLEMS IN CLEARANCE LEVEL INSPECTION AND SOLUTION USING NEW WASTE MONITOR TO ENSURE SOCIAL RELIANCE ON METAL RECYCLED FROM NUCLEAR FACILITIES

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ABSTRACT

The most important problem in the inspection of clearance level is how to ensure the social reliance on recycled metal from nuclear facilities. It is a serious concern that the circulation of recycled metal might be hindered if post-clearance-level-inspection metals were to develop a poor reputation. In order to remove this anxiety and ensure social reliance on recycled metal after inspection, the possibility of not detecting hot spots of contamination exceeding the surface contamination level must be eliminated completely. It also has to be clarified how the uncertainty of measurement results should be handled. To solve the above problems, a new probabilistic approach of establishing an appropriate safety factor for determining the uncertainty of measurements and nuclide spectra has been proposed. A new technique for precise and automatic measurements of gamma emitters in metal waste, the clearance automatic laser inspection system (CLALIS), has also been developed, which uses 3D noncontact shape measurement and Monte Carlo calculation techniques in order to objectively confirm that the specific radioactivity level of metal waste satisfies the clearance level and furthermore that its surface contamination level meets legal standards.

INTRODUCTION

The Nuclear Safety Commission in Japan (NSC) reported clearance levels for solid materials from nuclear reactors such as light water reactors (LWRs) and gas cooled reactor (GCR), in March 1999 [1]. Clearance level is the radioactivity level on the basis of which solid materials are or are not treated as radioactive waste. Clearance levels for nine important nuclides, shown in Table I, were presented in the NSC report. If the clearance levels are enforced under the present laws, concrete and metal wastes from decommissioned and active nuclear power plants could be disposed of as general industrial waste or recycled.

Table I. Clearance Levels for Nuclear Reactors (Mar. 1999) (Bq.g^{-1})

Nuclides	H-3	Mn-54	Co-60	Sr-90	Cs-134	Cs-137	Eu-152 Eu-154	Total Alpha
Level	200	1	0.4	1	0.5	1	0.4	0.2

The International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (BSS) [2] specifies the basic international requirements for health protection against exposure to ionizing radiation and for the safety of radiation sources. The BSS defines the concepts of exclusion, exemption and clearance. The applications of these concepts are written in the safety guide, RS-G-1.7 [3].

In December 2004, the Special Committee on Radioactive Waste Disposal Safety and Decommissioning Safety under the NSC completed reexamination of the clearance levels comparing the values of activity concentration derived using the exemption concept in RS-G-1.7. It was concluded that since there is no significant difference between the NSC levels and RS-G-1.7 values, it would be appropriate to adopt the RS-G-1.7 values as Japanese clearance levels from an international harmonization standpoint.

The basic concept of inspecting whether or not the radioactivities of concrete and metal wastes are below clearance levels was given in an NSC report presented in July 2001 [4]. In the inspection report, a simple method of inspecting clearance level was described. This method involves estimation of the radioactive concentration of all nuclides using the concentrations of key nuclides such as Co-60, which can be measured easily and is the dominant nuclide component. For nuclides whose concentrations cannot be measured by gamma measurement, an estimation method using previously assessed information on nuclide spectra and measurement results for a key gamma nuclide (hereinafter termed the nuclide spectrum (NS) method) can be applied.

In this paper, some problems regarding clearance level inspection have been listed. To solve these problems, a new approach of establishing an appropriate safety factor for determining the uncertainty of measurements and nuclide spectra has been proposed, which was adopted in the draft of Standard for Methods of Clearance Level Verification prepared by the Standards Committee (SC) of the Atomic Energy Society of Japan (AESJ). A new technique for precise and automatic measurements of gamma emitters such as Co-60 in metal waste has also been described as the solution.

PROBLEMS IN CLEARANCE INSPECTION

Uncertainty of Measurements and Nuclide Spectra

RS-G-1.7 provides us with activity concentration levels that are equivalent to clearance levels and also shows that the summation (hereinafter termed $\Sigma C/CL$) of the estimated nuclide concentrations (C) divided by each clearance level (CL) should be lower than 1 to satisfy the clearance criterion for a mixture of nuclides. It is also stated that the verification of the levels should be based on a procedure that may include direct measurement of materials, laboratory measurements of representative samples, and the use of properly derived radionuclide relationships.

As seen in RS-G-1.7 and a Japanese inspection report, the estimation of target nuclide concentrations for clearance inspection is based on the direct measurement of the concentrations of readily monitored nuclides, but for nuclides whose concentrations cannot be measured by gamma measurement, the NS method can be applied. As for target nuclides unsuitable for the NS method, the determination of their mean concentration is assumed as a conservative means of clearance inspection (hereinafter termed the mean concentration (MC) method).

In the case of clearance inspection, a low radioactivity results in many uncertainties (errors) of measurement. On the other hand, there may be a serious uncertainty (scattering) beyond the order of the activity concentrations and nuclide spectra used in the clearance judgment. These concerns cause a problem in determining how the uncertainty of measurements and the nuclide spectra should be handled.

Surface Contamination Measurement

In the inspection report [4], it was clarified that the surface contamination density of the inspection target must not exceed the surface contamination standard specified in Japanese law when the inspection target material is removed from a controlled area. Therefore an inspection of the contamination density (4 $Bq.cm^{-2}$ for beta or gamma emitters and 0.4 $Bq.cm^{-2}$ for alpha emitters; e.g., 100 cm^2 for averaging unit) is also necessary in the case of surface contamination. In the case of the inspection of the clearance level, a few tons of material can be treated as the averaging unit, and it is easy to measure high-level radioactivity if the mass of the measurement target is sufficiently large. On the other hand, in the case of the inspection of the surface contamination standard, the fact that the radioactivity level must be proven to be less than approximately 400 Bq for every 100 cm^2 of material presents a difficult problem.

Self-Shielding Effect for Gamma Measurement

Metal waste mainly originates from equipment in buildings, unlike concrete which is used as a structural building material. Since contaminated equipment must be decontaminated after dismantling, the main measurement targets are the fragments of equipment of various shapes, numbers and sizes. In the case of inspecting such non- uniform metal waste using a surface contamination survey meter or a surface contamination monitor that mainly measures beta rays, internal contamination such as activated contamination cannot be detected. In addition, there is a possibility of not detecting the contamination or underestimating the radioactivity level because the detection surface may not be close to the contaminated surface due to the complex shape of the measurement target. Moreover, in the case of internal contamination of a small metal pipe, measurement is impossible.

Not only beta rays but also gamma rays are emitted from Co-60, which is the dominant nuclide in metal wastes from decommissioned and active nuclear power plants. Gamma ray measurement is suitable for such internal contamination as mentioned above because of the strong penetrative ability of gamma rays. For gamma ray measurement, accurate and easy calibration of the actual radioactivity level and count rate obtained by measurement is important. If gamma ray measurement can confirm that the radioactivity level is less than approximately 400 Bq, both clearance level and surface contamination level can be inspected simultaneously. In addition, the large amount of effort required for manual inspection using a survey meter could be minimized, and nondetection of hot spots of radioactivity due to human error would not occur.

For the inspection of clearance level for large metal waste, a gamma monitor having a measurement container such as a basket with some large plastic scintillation detectors is used. When using such a gamma monitor, the measurement of low-level radioactivity without compensating for gamma shielding effects due to the presence of the metal itself causes an underestimation of radioactivity, as pointed out in an important warning item in ISO11932 [5], the international standards for radioactivity measurements of clearance level. In order to overcome such a problem, it is necessary to perform many calibrations of the filling density of a measurement container for every type of metal waste, but these calibrations require much effort.

There is also the problem that the human factor involved in the selection of a suitable calibration factor for the measurement target from a calibration database may result in a lack of objectivity and a decrease in the reliability of the measurement data.

Reliance on Measurement

The most important problem in the inspection of the clearance level is how to ensure social reliance on recycled metal from nuclear facilities. Metals after inspection could be disposed of as general waste. However, from the viewpoints of reducing environmental burden and effective utilization of resources, recycling should be the primary objective for metals after inspection. At present, the recycling of metal is proceeding well in Japan. However, it is a serious concern that the circulation of recycled metal might be damaged if the post-clearance-level-inspection metals were to develop a poor reputation. In order to remove such anxiety and ensure social reliance on recycled metal after inspection, the possibility of not detecting hot spots of contamination exceeding the surface contamination level must be eliminated completely.

In selecting the measurement tools and the measuring conditions, it should be noted that detection limit has to be estimated appropriately considering the uncertainty in correcting gamma shielding effects due to the presence of the metal itself and calibration error. To achieve a modified detection limit of a few 100 Bq, it has already been clarified in the previous paper [6] that the calibration error and the correction error caused by the self-shielding effects of background count rate have to be limited within a few tens and a few percentages, respectively, and that a long measurement time is not effective.

SOLUTION FOR UNCERTAINTY

Concepts of Dose Criterion of Clearance Level

In the derivation of clearance level in Japan, the dose criterion for clearance was of the order of 10 μSv/y, which was obtained by reducing 100 μSv/y, which is equivalent to a negligible risk level (10^{-6} /y) that nobody considers in determining their action to one tenth while considering the overlap of practices and exposure pathway. In addition, using a stochastic approach, the 97.5 percentile of the dose distribution, which is equivalent to the higher endpoint of the 95 % confidence interval, was confirmed to be lower than 100 μSv/y for all exposure scenarios. Thus, the clearance level of solid materials is not strictly required to be lower than 10 μSv/y in Japan. In the BSS, the dose criterion is also described as being of the order of 10 μSv or less in a year, but is not exactly 10 μSv/y. The concept of the dose criterion of clearance level in Japan is consistent with that specified by the BSS.

This indicates that the clearance dose level for solid materials may be stochastically permitted to be higher than 10 μSv/y. Therefore, for clearance level inspection, it is against the basic concepts of the dose criterion for clearance to choose a safety margin for clearance judgment in order to strictly maintain 10 μSv/y.

Conservativeness Involved in Clearance Judgment

The clearance level in Japan was derived for every nuclide and determined for the most serious exposure pathway. To determine whether $\Sigma C/CL$ is lower than 1 requires us to consider some rare cases in which the most serious scenarios with different nuclides will occur simultaneously. This conservativeness in the overlap of practices and exposure pathways in clearance judgment using the relation $\Sigma C/CL<1$ is considered when a dose criterion of 10 μSv/y is set by reducing 100 μSv/y, which is equivalent to a negligible risk, to one tenth. This implies a double consideration of the overlap of practices and exposure pathways. In RS-G-1.7, clearance judgment using the relation $\Sigma C/CL<1$ was also required for a mixture of nuclides, which means the consideration of rare cases in Japan.

From the above-mentioned reason, it can be concluded that a safety margin is involved in clearance judgment using the relation $\Sigma C/CL<1$.

Basic Concepts of Safety Margin in Clearance Judgment

Taking the basic concept of the dose criterion for clearance and the conservativeness involved in the clearance judgment into consideration, the following concepts of safety margin for clearance have been adopted in a draft of the Standard for Clearance Level Inspection to be released by the SC of AESJ.

a) When the 97.5 percentile of the probability distribution of $\Sigma C/CL$ due to the uncertainty of measurement of a key nuclide, previously assessed mean concentrations and concentration ratios of the other nuclides to the key nuclide, is ten times higher than the $\Sigma C/CL$ obtained using a geometric mean of C, a factor of (97.5 percentile / 10) is required for the safety margin in clearance judgment.

b) When the 97.5 percentile of the probability distribution of $\Sigma C/CL$ due to the uncertainty of measurement of a key nuclide, previously assessed mean concentrations and concentration ratios of the other nuclides to the key nuclide, is NOT ten times higher than the $\Sigma C/CL$ obtained using the geometric mean of C, no safety margin is required in clearance judgment.

Method of Calculation of Probability Distribution of $\Sigma C/CL$

The treatment of the uncertainty of measurement of a key nuclide is deeply related to the concept of detection limits. The uncertainty of measurement is generally expressed as a normal distribution. In Japan, the detection limits for radiation measurements are defined as 3σ, which is three times the standard deviation of the measurement results. In the case of the monitor checked on such a detection limit, the relative error of measurement results is always less than approximately 30 % since the measurement results are usually beyond the detection limit. This indicates that an uncertainty of more than approximately 30 % is not required in the measurement results.

In the U.S., the concept of detection limit is expressed by the minimum detectable amount (MDA), as reported by Currie [7]. In this case, the detection limit cannot simply be expressed by a factor of the standard deviation, but is approximately regarded as 3.29σ, which is twice the value of 1.645σ. This implies that an uncertainty of more than approximately 30.4 % is not required in the measurement results, which is the same conclusion derived from the Japanese concept of detection limit.

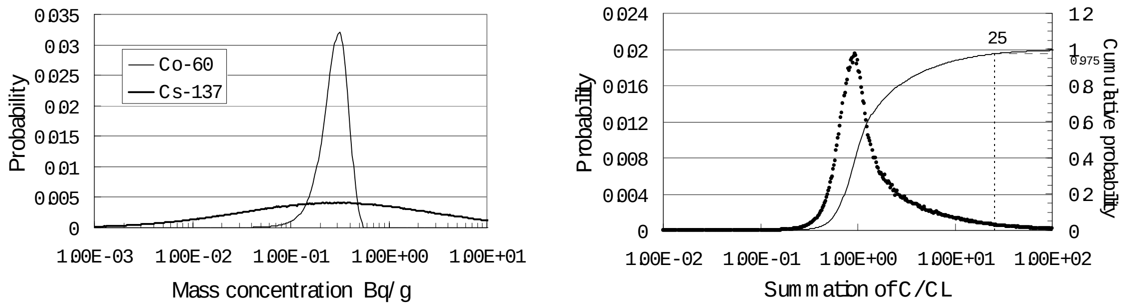
On the other hand, there may be extensive scattering beyond the order of mean concentration (MC method) or concentration ratios of the other nuclides to the key nuclide (NS method). The frequency distribution of the activity concentrations and nuclide ratios can be generally expressed by a log normal distribution and determined using two parameters, the geometric mean and geometric standard deviation. To estimate the uncertainty of the mean concentrations and concentration ratios of the other nuclides to the key nuclide, one must know the geometric standard deviation that indicates the degree of scattering, after the confirmation of whether the frequency distribution can be fitted to the log normal one.

If the uncertainty of the measurement of the key nuclide, the mean concentrations and concentration ratios of the other nuclides to the key nuclide can be simultaneously considered in the calculation of the probability distribution of $\Sigma C/CL$, their separate safety margins are not required. If a relative measurement error of 30 % is used in the calculation of $\Sigma C/CL$, different safety margins for every measurement are not required.

From the above results, to practically use the basic concepts of the safety margin for clearance judgment, the probability distributions of $\Sigma C/CL$ have to be calculated by assuming that the uncertainty of the measurement results can be expressed as a normal distribution with a 30 % relative error that is equal to the coefficient of variation, and the uncertainty of the mean concentrations and concentration ratios of the other nuclides to the key nuclide can be treated as log normal distributions. The calculations of the normal distributions can be theoretically treated in a mathematical way, but the Monte Carlo calculation is the most suitable for such a calculation of the summation of the normal and log normal distributions.

Using the Monte Carlo calculation, the probability distribution calculation system (PDCS) has been developed, which can calculate the probability distribution of $\Sigma C/CL$ by taking the uncertainty of the measurement results, mean concentrations and concentration ratios into account. PDCS can determine the probability distribution of $\Sigma C/CL$ with normalized $\Sigma C/CL$ obtained by giving C a geometric mean. In PDCS, a key nuclide has to be freely selected from the target nuclide. Both geometric mean and geometric standard deviation have to be given for the mean concentration and nuclide ratio.

As an example of the use of PDCS, the probability distribution of $\Sigma C/CL$ was calculated. Figure 1a) shows sample input data. The key nuclide used for measurement was Co-60 and the uncertainty was assumed to be 30 %. The nuclide assessed by the NS method was Cs-137. The geometric mean and geometric standard deviation of Cs-137/Co-60 concentration ratio were 1.0 and 9.6, respectively. The concentration of Co-60 was normalized so as to satisfy the relation $\Sigma C/CL=1$. Figure 1b) shows the output data of the probability distribution of $\Sigma C/CL$. It can be seen from Fig. 1b) that the 97.5 percentile of the probability distribution of $\Sigma C/CL$ is 25. According to the basic concepts of the safety margin for clearance judgment, a factor of 2.5 (=25/10) for the safety margin is required in this case. That is, if there are many uncertainties in the Cs-137/Co-60 concentration ratio, it would not be sufficient for clearance to confirm the relation $\Sigma C/CL<1$. The verification of $\Sigma C/CL<0.4$ (=1/2.5) would then be required for clearance judgment.



a) Input data b) Output data

Fig.1 Sample data for PDCS

SOLUTION FOR MEASUREMENTS

Clearance Automatic Laser Inspection System, CLALIS

To reduce the possibility of not detecting hot spots of contamination exceeding the surface contamination level, gamma measurement is available. The application of gamma measurement to surface contamination survey requires a high detection sensitivity of less than a few 100 Bq for all the surface of the measurement target. To realize this, the detection limit has to be improved appropriately considering both uncertainties of the calibration factor and the correction factor for self-shielding effects. To decrease the two uncertainties, objectivity should also be ensured while reducing the human factor as much as possible.

To satisfy such requirements, a practical monitor (clearance automatic laser inspection system, CLALIS) has been developed as shown in Fig. 2. In the monitor, 1-200 kg of metal waste, which is less than the measurement volume of 80 cm x 80 cm x 40 cm (height), can be treated using 4 laser scanners. A value of 15 % for the minimum efficiency at any point in the measuring volume can be obtained using eight (4 upper and 4 lower) large plastic scintillation detectors.

Fig.2 Appearance of CLALIS.

The flowchart of the system is shown in Fig. 3. First, after mass measurement, the shape of the metal waste is measured using a noncontact shape measurement system with a laser, then information on the locations of the measurement target and the plastic scintillation detector is obtained by dot imaging. Secondly, the location of the metal waste is transferred to the gamma monitor; during the transfer, two calculations (MCNP Ver.4C) are carried out based on the observed dot image.

The first MCNP calculation is for the correction of the gamma shielding effect due to the presence of the metal waste itself. The decrease in background count rate results in the radioactivity level being underestimated or the radioactive contamination not being detected. In this study, this difficult problem was solved by the development of an automatic correction method for background count rate. The outline of the method is as follows.

Based on the shape measurement data of the metal waste, the efficiencies of gamma rays emitted from the surface surrounding the gamma monitor are calculated by the MCNP code for two cases: the case in which the measurement target is in the gamma monitor and that in which there is no target. The ratio of the two efficiencies is the correction factor. The automatic correction is completed multiplying the actually observed background count rate by the correction factor. The exact net count rate can be obtained by subtracting the corrected background count rate from the total one.

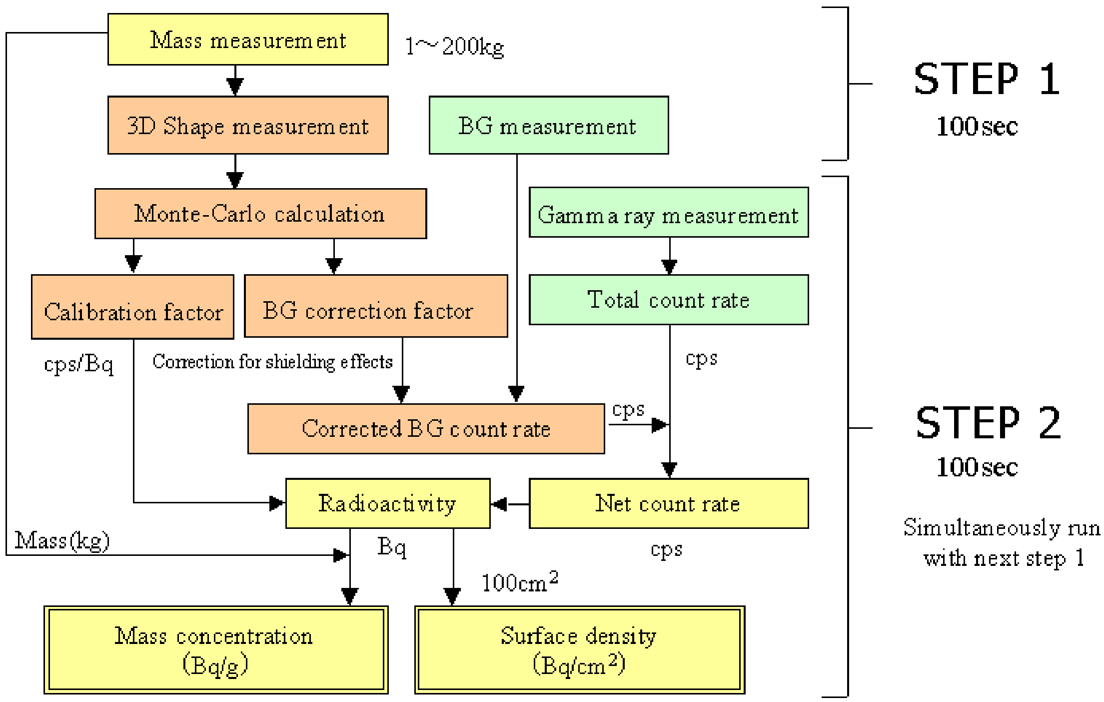


Fig. 3 Flowchart of CLALIS.

The second MCNP calculation is for the calibration factor. The calibration factor can be obtained by calculating the efficiency of gamma rays emitted from the measurement target and converting it into the calibration factor ($cps.Bq^{-1}$, count rate per radioactivity) using the gamma ray emission probability per disintegration of Co-60. In the case of the inspection of uniformly distributed activated contamination, gamma rays are emitted uniformly from inside the target. On the other hand, for local distributions such as surface contamination, hot spots are automatically selected in the location of minimum sensitivity so as to give a conservative estimation.

The radioactivity of the measurement target is obtained by dividing the net count rate by the calibration factor. The measurement for the clearance level is carried out based on the mass concentration obtained by dividing the radioactivity by the actual measured mass. In the case of the measurement for the surface contamination level, surface density is calculated by dividing the radioactivity by a fixed value of 100 cm^2 based on Japanese Industrial Standards (JIS) Z 4504. This estimation is conservative so as not to exceed the legal level of surface contamination density for every 100 cm^2 of the entire metal surface. This makes it possible to minimize effort for a 100 % manual survey and to remove the possibility of not detecting hot spots at locations that cannot be scanned easily with a survey meter.

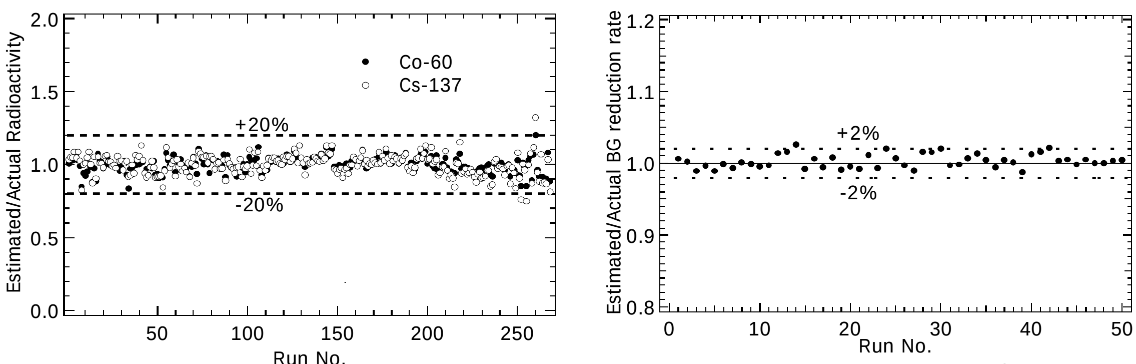
Measurement data can be obtained with a high objectivity independent of the skill of the workers because all calculations are automatic. The accuracy of the measurement is high because calibration and background correction are performed for every measurement target.

Accuracies of Calibration and Background Correction

To investigate the calibration error of CLALIS in the case of local contamination, the results of measurement using the monitor were compared with the known radioactivities of Co-60 and Cs-137 sources, using 268 mock metal waste samples of various shapes, sizes and numbers [8]. The gamma sources were located at positions where the sensitivity of the monitor was minimum. The mock metal waste samples were as follows: JIS 6A, 20A, 40A, 65A, 100A and 250A SUS pipes with lengths of 10, 20, 30, 40 and 80 cm, SUS 20 x 10 cm plates with thicknesses of 1, 2, 3, 4, 5 and 6 cm, and SUS valves with complex shapes.

The result of the comparison is shown in Fig. 4a). The data measured using the present monitor and actual radioactivity levels were in good agreement within +/- 20 %. The calibration error can be estimated to be approximately 7 % using the standard deviation of all the data. In the case of a uniform distribution, such as in activated contamination, a similar comparison using MCNP calculations was carried out, which showed almost the same results as those for the local contamination cases.

The accuracy of the correction error of background reduction was investigated using the 50 samples selected from the above 268 samples as marked background reduction cases. The results are shown in Fig. 4b). It can be seen that the corrected and actual background reductions were in good agreement within +/- approximately 2 %. The correction error of background reduction can be estimated to be approximately 1 % using the standard deviation of all the data.



a)

Calibration errors b) Correction errors for BG reduction

Fig. 4 Accuracies of calibration and correction for background reduction.

As for the detection limit, Currie describes in a previous paper [7] that, in the United States, the minimum detectable amount (MDA), defined in Currie's paper as the 'detection limit', is usually used as the detection limit [9,10]. The MDA of the count rate is given by

$$n_{MDA} = \frac{2.71 + 4.65\sqrt{n_B t_B}}{t_T}, \qquad \text{(Eq. 1)}$$

where

n_{MDA} Detection limit of the count rate (sec^{-1});

t_T Measurement time (sec);

t_B Measurement time for BG (sec);

n_B Background count rate (sec^{-1}).

Generally in Japan, the detection limit of the count rate is given by

$$n_D = \frac{3}{2}\{\frac{3}{t_T} + \sqrt{(\frac{3}{t_T})^2 + 4n_B(\frac{1}{t_T} + \frac{1}{t_B})}\},\qquad \text{(Eq. 2)}$$

as shown in the inspection report [4]. n_D is also the detection limit of count rate, and Eq. 2 is qualitatively equivalent to Eq. 1. Equation 2 is defined in Currie’s paper as the ‘determination limit’. When the calibration error and the BG correction error are taken into account in the detection limit evaluation [6], Eq. 2 can be expanded to

$$A_{DL} = CF\,\frac{\frac{3^2}{t_T} + \sqrt{(\frac{3^2}{t_T})^2 + 4(1-3^2 r_2^2)3^2\{n_B(\frac{1}{t_T} + \frac{1}{t_B}) + r_1^2 n_B^2\}}}{2(1-3^2 r_2^2)}.\qquad \text{(Eq. 3)}$$

here,

A_{DL} Detection limit of radioactivity (Bq);

CF Calibration factor (i.e. Bq. sec);

r_1 Relative error of BG correction;

r_2 Relative error of calibration.

By substituting 7 % of the calibration and 1 % of the background correction errors into Eq. 3, the detection limit of CLALIS was estimated. A value of 100 Bq for the detection limit was obtained for Co-60 in the case of a 100 sec (30 sec gamma ray measurement, 30 sec background measurement) process time per inspection. This indicates that 5 tons of metal waste per day (1,000 tons per year) can be measured in batches of more than 20 kg at that process speed.

CONCLUSION

As a solution for the treatment of an uncertainty in clearance inspection, a probabilistic approach to providing an appropriate safety margin for clearance judgment has been proposed taking the concepts of the dose criterion of clearance level and conservativeness involved in the clearance judgment into account. To practically apply the concepts to clearance judgment, a probability distribution calculation system (PDCS) has been developed, which can calculate the probability distribution of ΣC/CL while taking the uncertainty of the measurement results, mean concentrations and concentration ratios into account.

A new monitor for precise and automatic measurements of gamma emitters in metal wastes, clearance automatic laser inspection system (CLALIS) has also been developed using 3D noncontact shape measurement and Monte Carlo calculation techniques, in order to objectively confirm that the specific radioactivity level of metal waste satisfies the clearance level and furthermore, that the surface contamination level of the metal waste is below the legal standard level. From the results of the performance tests of the present monitor, it was clarified that a detection limit lower than 100 Bq was achieved by taking into account the calibration error and correction error of the shielding effects of metal. This indicates that the present monitor can also realize a practical process speed.

The CLALIS is a new measurement monitor that can detect a low level of radioactivity such as 100 Bq. The judgment method for the satisfaction of legal surface contamination level using the monitor is more conservative than the manual survey method for every 100 cm^2 area, which may lead to the absence of contamination in the entire measurement target; however, the present monitor can greatly reduce the amount of effort required in carrying out a 100 % manual survey of metal surfaces, provide objectivity for inspection data, and completely eliminate the possibility of not detecting hot spots of radioactive contamination.

The measurement target for inspection by CLALIS is now limited only to metal waste generated at the early stage of the decommissioning of a nuclear power plant. A further study on the application of this method to concrete waste including natural radionuclide has already been planned. We hope this system will actually be used in the decommissioning of nuclear power plants in the near future.

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