PRACTICABLE ASSAY SYSTEM FOR RADIONUCLIDE QUANTIFICATION OF DISPOSAL PACKAGES

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ABSTRACT

The requirements for land disposal of LLW from nuclear power plants and facilities have necessitated a more complete and accurate analysis of the radionuclide contents of waste packages.

JGC has developed a new direct assay technique based on gamma-ray spectroscopy and total gamma-ray counting, combined with a scaling factor methodology for difficult-to-measure nuclides.

The system consists of an HpGe detector, a plastic scintillator, a microcomputer and a waste package handling system such as a turntable.

The radioactivity concentrations of Co-60 and Cs-137, which are key nuclides for difficult-to-measure nuclides, are calculated from the activity ratio of Co-60 to Cs-137 measured by using an HpGe detector and total radioactivity is measured by using a plastic scintillator.

The concentrations of difficult-to-measure nuclides in a waste package are calculated by combining the radioactivity concentrations of Co-60 and Cs-137 with the waste package data and scaling factors.

The system is simple and enables a complete analysis of all nuclides specified prior to shipment and disposal of waste packages, and also ensures that not only homogeneous solidified waste but also nonhomogeneous DAW (dry active waste) can be measured within a short time.

INTRODUCTION

In land disposal of radioactive solid waste generated at nuclear power plants, it is necessary to assay radionucludes contained in waste packages for the safe operation of the disposal site.

Important nuclides from the standpoint of land disposal are Co-60 and Cs-137.

In addition, difficult-to-measure nuclides such as C-14, Ni-63, Sr-90, etc. are also listed (see Table I).

In shipping waste packages from nuclear power plants to the disposal site, confirmation of the contents of these nuclides is also required in compliance with disposal package technical package requirements.

Considering such background needs, JGC has been developing the most suitable radionuclide assay technique for nuclear power plants since 1983 and has developed a new radionuclide assay system which combines the scaling factor method and the gamma scanning and gross gamma counting method.

This radionuclide assay system is essentially simple in nature. It is capable of counting the radioactivity of not only homogeneous solidified waste but also nonhomogeneous waste within a short time, and due consideration has been given to the suitability of the confirmation techniques applied in waste package shipment.

The principle, composition, performance, etc. of the radionuclide assay system based on the gross gamma method are explained below.

SYSTEM CONCEPT

In order to assay the radioactivity content specific to nuclides in shipping waste packages from nuclear power stations, a radionuclide assay system suitable for this purpose must be developed with consideration given to the following matters.

- Capable of assaying α and β nuclides which are important in land disposal.
- Applicable to waste which is varied in type, size, and weight.
- Capable of covering the measurement of extensive radioactivity concentrations (most DAW is distributed in the extremely low level region).
- Simple system and short measuring time.

To solve these problems, JGC has developed a new radionuclide assay system based on the following approaches.

- Evaluation of the correlation between difficult-to-measure nuclides and key nuclides and data verification.
- Simple nondestructive direct measurement of key nuclides.

TABLE I

Activity Concentration Limits of LLW Burial in Japan

(γCi/g)

C-14	1×10^{0}
Co-60	3×10^{2}
Ni-63	3×10^{1}
Sr-90	2×10^{0}
Cs-137	3×10^{1}
TRU	3×10^{-2}

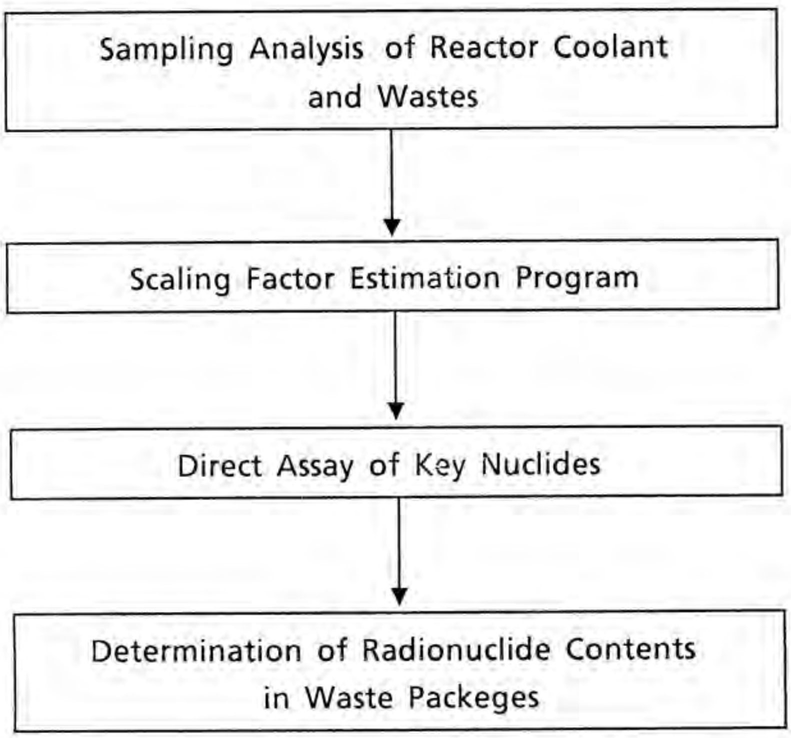

Fig. 1. Concept of Assaying Radionuclides in Waste Package.

As shown by Fig. 1, the former approach has been attained basically by establishing a scenario for nuclide behavior in nuclear reactor systems and determining the correlation between key nuclides and difficult-to-measure nuclides (C-14, Ni-63, Sr-90, TRU nuclides, etc.).

The latter approach is based on the method in which the radioactivity of key nuclides is assayed by selecting Co-60 as a key nuclide for CP nuclides (and C-14, TRU nuclides) and Cs-137 as a key nuclide for FP nuclides from among the nuclides important from the standpoint of land disposal, then measuring the ratio between Co-60 and Cs-137 and the total gamma radiation. (Co-60 is used as an empirical key nuclide for C-14 and Cs-137 for TRU nuclides.)

This system is intended to be used for the nondestructive inspection of waste packages before shipment. Therefore, in settling its target performance, due consideration must be given to the trend of applicable laws and regulations, and the actual conditions of the waste.

Trial calculation results have been obtained which indicate that if the radioactivity concentrations of Co-60 and Cs-137 can be measured to 10^{-4} μCi/g or so, there is no effect on the disposal capacity which is important in safety evaluation, when the trend of applicable laws and regulations up to the present is considered.

From the conditions of actual waste at nuclear power plants, it is found that especially the radioactivity concentration of DAW is distributed in a wide range centering around the low concentration region. Therefore, measures to cover every type of waste package are required and the range of the relative concentration ratio between Cs-137 and Co-60 must be considered.

Considering these requirements, the target performance of our system is as follows.

Measuring range	: 10-5 - 10^{-1} μCi/g
Measuring Cs-137/Co-60 ratio	: 1/100
Measuring time	: 5 minutes

BASIC PRINCIPLES

As shown by Fig. 2, in this nuclide assay system, the concentrations of radionuclides contained in waste packages are estimated by measuring the radioactivities of Cs-137 as a key nuclide for FP nuclides and Co-60 as a key nuclide for CP nuclides and considering scaling factors, half-lives, etc.

The gamma-ray counts from any point inside a drum of waste can be express by the following equation on condition that no consideration is given to the background gamma-ray.

$$\sum_{i=1}^{n} Ni(E) = \varepsilon(E) \sum_{i=1}^{n} Si(x,y,z,E) \cdot \frac{\Omega i(x,y,z)}{4\pi} \cdot \exp(-bi(x,y,z,E)) \quad (1)$$

Where, Ni(E) : Detector count of the radiation from the radiation sources Si

ε(E) : Detection efficiency for each energy

Si(x,y,z,E) : Intensity of the radiation source inside the drum

Ω i(x,y,z) : Detector solid angle as seen fromthe radiation source Si

bi(x,y,z,E) : Attenuation distance from radiation source Si to the detector

Eq. (1) indicates that the gamma-ray counts at the detection point depends on the geometrical efficiency at any point inside the package and the attenuation distance between the radiation source and the detector.

In this system, the geometrical efficiency at any point inside the drum is made constant by measuring the gamma-ray flux radiated from the whole circumferential surface of the drum and correction for attenuation is made based on the mean density of the waste (weight/volume).

Thus, Eq. (1) is expressed as follows.

$$\sum_{i=1}^{n} Si(x,y,z,E) = \frac{\sum_{i=1}^{n} Ni(E)}{\varepsilon(E) \cdot \frac{\overline{\Omega i}}{4\pi} \cdot \exp(-\overline{bi}(E))} \quad (2)$$

As the measured value Ni(E) is a function of energy, it is necessary to identify the nuclides to be measured.

Assuming that the nuclides to be measured are Co-60 and Cs-137:

$$\Sigma Si\ {}^{60}Co = \frac{\sum_{i=1}^{n} Ni({}^{60}Co)}{\varepsilon({}^{60}Co) \cdot \frac{\overline{\Omega i}}{4\pi} \cdot \exp(-\overline{bi}({}^{60}Co))}$$

$$\Sigma Si\ {}^{137}Cs = \frac{\sum_{i=1}^{n} Ni({}^{137}Cs)}{\varepsilon({}^{137}Cs) \cdot \frac{\overline{\Omega i}}{4\pi} \cdot \exp(-\overline{bi}({}^{137}Cs))} \quad (3)$$

As the plastic scintillator is incapable of spectroscopic analysis, it is impossible to distinguish Co-60 and Cs-137 from each other. Therefore, the Cs-137/Co-60 ratio must be measured using another detector. This system uses a Ge semiconductor detector for this purpose.

In this system, in order to make the geometrical efficiency constant, the mean geometrical efficiency is raised

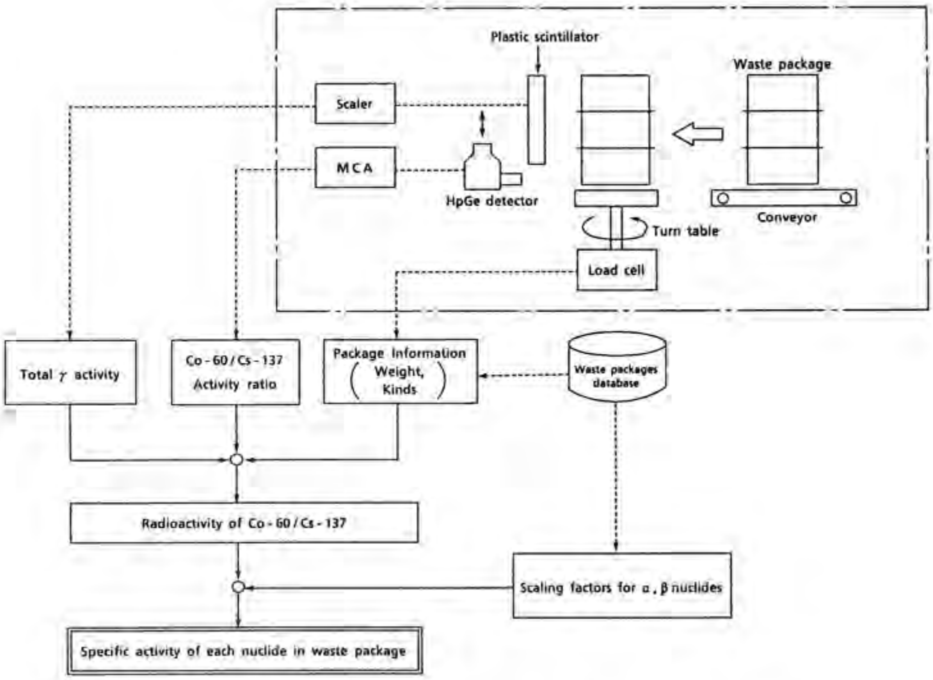

Fig. 2. Schematic Diagram of Nuclide Assay System

to 0.5 - 0.6 by rotating the drum and disposing the plastic scintillator so as to cover the overall height of the drum.

APPLICATION TO WASTE PACKAGES

As described above, the radiation incident on the scintillator from the sources contained in the waste package is expressed by Eq. (1) and varies in accordance with the location of the sources and the effects of the attenuation depending on the waste type.

In order to ensure accurate measurements, it is therefore necessary to make corrections after grasping the condition of the content of the package or to devise a measurement method which is not easily affected by the condition of the content.

In the case of homogeneous solidified waste, the density of the content and the distribution of radioactivity are uniform. Therefore, once the solidifying agent used and the target nuclides are determined, the variables used in the equations can be made constant and accurate measurements can be ensured.

In the case of DAW packages, however, the kind of waste, packing condition, contamination pattern, etc. are varied and the measured values are affected by these conditions.

The effects of these conditions are observed as the attenuation of radioactivity in waste packages. For instance, the measured values become large when nuclides exist near the drum surface and become small when they exist in the center of the drum.

Such difference in the measured values becomes larger as the density of the content of the drum becomes larger.

In this system, the amount of radioactivity is counted as the gross gamma radiation dose whose energy region is not specified.

Radiation is attenuated due to the scattering of radiation by the contents of packages, but in this system, such scattered rays are also counted together.

It is known that the degree of scattering tends to increase as the density of the content becomes higher and as the transmission distance in materials becomes longer.

Therefore, even if materials having a large attenuation effect unevenly exist in the drum, the influence of such materials can be averaged by measuring the gross gamma radiation dose from various directions but not from a particular direction.

As described above, this system is aimed at correcting measurements for the effects of the uneven distribution of density and radionuclides in waste packages and also improving the measuring time and the detection limits by providing several detectors.

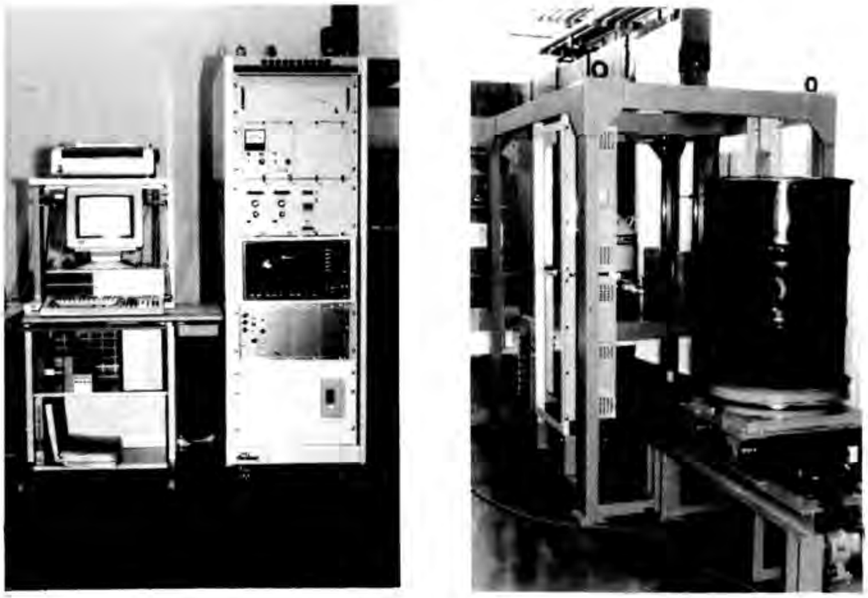

Fig. 3. Photograph of Prototype Assay System.

TABLE II

Basic System Specifications.

Item	Specifications
Waste packages to be measured	200 - ℓ standard steel drum Package
Nuclides to be measured	· Directly measured nuclides : Co - 60 and Cs - 137 , etc. · Derivatively measured nuclides : Sr - 90 , Ni - 63 , etc.
Measurable activity	Approximately 0.1 μ Ci / drum (Co - 60 / Package Density : 0.5)
Measuring time	5 min / drum
Detectors	· High - purity Ge semiconductor detecor for measurment of Cs - 137 and Co - 60 · Plastic scintillator for measurement of the gross γ radiation dose
Operation and Control	· Automatic computer control

BASIC SYSTEM SPECIFICATIONS

The basic system specifications are shown in Table II and each item is detailed below. A photograph of the prototype assay system is shown in Fig. 3.

1. Waste packages to be measured

The standard system is calibrated to measure the radioactivity of 200-liter drum waste packages. The system is also applicable to waste packages of other drum sizes by being calibrated. Concerning the kinds of waste to be measured, the system is also applicable to homogeneous solidified waste and DAW.

2. Nuclides to be measured

In principle, the nuclides to be measured are Co-60 and Cs-137, which are the key nuclides in the scaling factor method. However, an optional device permits the measurement of the key nuclide concentration even if waste packages specific to sites contain interfering nuclides such as Mn-54.

The concentrations of difficult-to-measure nuclides such as Ni-63, Sr-90, etc. are calculated based on the data base, using the scaling factor method.

3. Measurable activity

The system is capable of measuring the radioactivity up to approximately 0.1 μCi/drum as gamma-ray radioactivity (for Co-60 when the bulk density of waste packages is 0.5).

4. Measuring time

The standard measuring time is 3 minutes but selection can be left to the operator's selection.

Data processing requires about 2 minutes.

5. Detectors

A high-purity Ge semiconductor detector is used to measure the Cs-137/Co-60 ratio, and a plastic scintillator is used to measure the gross gamma radiation dose.

6. Operation control

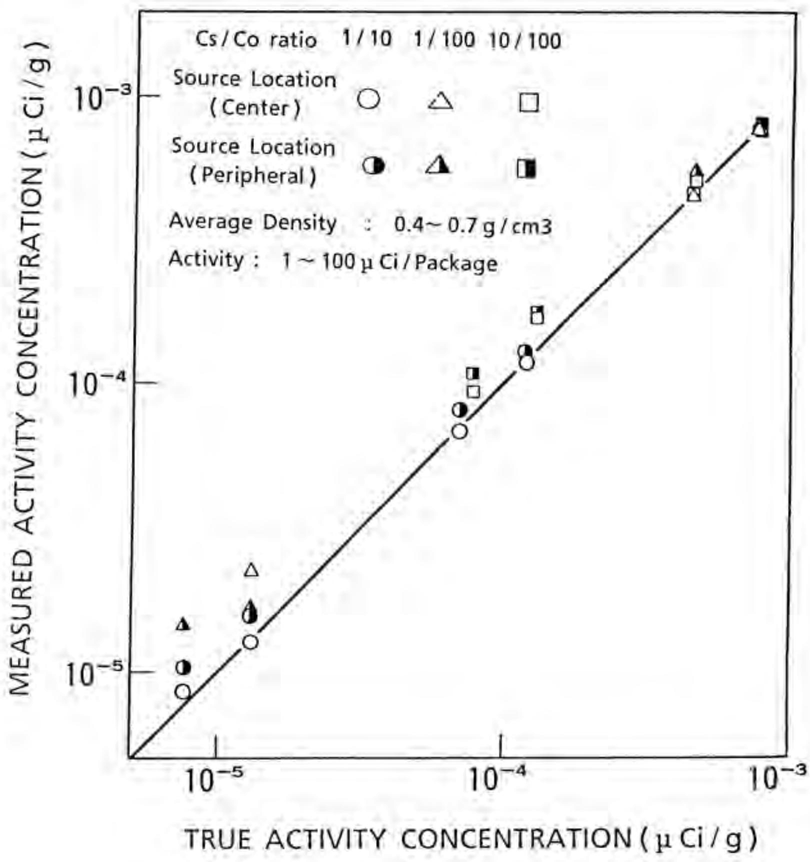

Fig. 4. Measurement Results of Pattern Test.

The operation of the system is automatically controlled by a computer. All waste package information can be inputted by interfacing with the computer using the keyboard. (The system can be connected to a superior computer system.) As-measured data, i.e., MCA spectrum data and scaler counts are stored in a floppy disk to permit easy retrieval of past measured data and easy erasure of unnecessary data.

SYSTEM PERFORMANCE

The performance of this system was evaluated stepwise firstly by a pattern test, then a full-scale simulated waste package test, and finally an actual waste package test. Especially in applying this system to DAW packages, such an approach is important because it is impossible to specify the geometrical configuration and the distribution of density and radioactivity in such a package.

An example of the results of each test is described below.

1. Pattern test

In this test, measurements were conducted by preparing a simulated waste package whose inside was divided into several segments so that the density of waste in each segment could be changed, and placing a standard radiation source in the specified position inside the package.

An example of the measurement results is shown in Fig. 4.

2. Full-scale simulated waste package test

In this test, more realistic measurements were conducted, using full-scale simulated waste packages similar to actual ones in the aspects of configuration and material, which were obtained by extending a model package. A single sealed radiation source was used in this test.

3. Actual waste package test

In this test, a reference simulated waste package was used for calibration purposes, and then performance tests were carried out by using actual waste packages (cement packages and DAW packages).

The tests on the cement packages were conducted to ascertain the calibration constant, and it was

confirmed that errors in the key nuclides measurements were within 5%.

The results of the measurements of the key nuclides were compared with the activity of difficult-to-measure nuclides, which was estimated by the scaling factor method and also with that of the core sample which was analyzed by a manual method.

The tests on DAW were implemented to check for errors due to changes in the filling pattern of radwaste, and it was established that the deviation factor was within $\pm 5\%$ at 1σ when the bulk density was 0.5 g/cm^3.

CONCLUSION

This system is very simple in mechanism and capable of assaying all radionuclides which are important from the standpoint of land disposal. In addition, it is widely applicable to not only homogeneous solidified waste packages but also nonhomogeneous DAW packages. Therefore, this system can be said to be most suited for nondestructive inspection required in shipping waste packages from nuclear power plants to disposal sites. In future studies, more extensive simulation tests and site verification tests will be continued further and the scaling factor estimation program will be improved.