

DEVELOPMENT OF NONDESTRUCTIVE MEASUREMENT TECHNIQUES FOR URANIUM-CONTAMINATED WASTE IN CONTAINERS

Y. Aoyama, K. Ohki
Japan Nuclear Cycle Development Institute

Y. Sukegawa, S. Suzuki
Nuclear Technology & Engineering Co., Ltd

H. Sagawa
Mitsubishi Heavy Industries, Ltd

H. Doi, Y. Endo
Nuclear Development Co.

ABSTRACT

We have developed a new technique based on the passive gamma method for measuring a quantity of uranium in a uranium waste container (1200 liter volume). The measurement system consists of three NaI (Tl) detectors, one High Purity Germanium (HPGe) detector and a handling system. The measuring method uses two discrete gamma-ray energies emitted from the daughter nuclide Pa-234m (1001 keV and 766 keV). The technique is applicable even if distribution of density and radionuclides are not uniform.

We developed a new technique to remove the influence of distances between radionuclides and detectors. This technique was named "the Facing Couple Method (FCM)".

If the influence of daughter nuclide is strong, the evaluation is carried out using data of detector to remove the influence. When counting rates are low, gross counting rates corrected the variation of background rate by container is adopted as an alternative method.

We confirmed applicability of this technique and system by examination using natural uranium (50g - 10kg) and the observation of container started from January 14, 2004 and 107 observations have been completed until August 23, 2004.

INTRODUCTION

Measurement evaluation of uranium content within wastes is necessary for waste management, waste treatment and disposal.

The majority of uranium wastes in "Japan Nuclear Cycle Development Institute (JNC)" are packaged in 200 liter drums or 1200 liter rectangle containers.

The quantity of uranium in drums has been measured by non-destructive assay (passive gamma method) in JNC (1) (2).

By in contrast, the quantity of radionuclides in the container is difficult to evaluate.

The measuring method of the waste drum with revolution cannot apply to the rectangle container.

One exception to this is a plutonium waste container.

The container contained plutonium was measured by “the Waste Container Assay System (WCAS)” in JNC.

The system utility of the WCAS is the passive neutron coincidence method (3).

However, the technique such as WCAS cannot apply to the uranium waste, because the uranium waste releases little neutron.

The estimation of quantity of uranium relied on waste information that composition of uranium and surface dose rate.

For the reasons mentioned above, we have developed the new technique based on the passive gamma method for measuring the quantity of uranium in the uranium waste container.

Waste and Container Characteristics

The uranium waste containers are loaded with a large variety of waste materials contaminated with uranium and its daughter nuclides.

For example, steel or PVC pipes, steel racks, pumps and etc. are packaged in a container. The distributions of density and radionuclides are not uniform.

The following is a physical description of the container.

The dimensions of a container are approximately 120 x 120 x 90 cm with each container likely to hold up to 1200 liter waste volumes.

A wall thickness of container is 0.45cm and that of bottom is 0.6cm of steel (SS400).

The weight of an empty container is 400 kg.

System

The measurement apparatus was manufactured in cooperation with Mitsubishi Heavy Industries, Ltd. The system was designed to assay containers with volume up to 1200 liter and weights up to 3000 kg in its standard configuration.

The measurement system consists of three collimated NaI detectors, one collimated HPGe detector and a handling system which could move a container back and forth and turn it (Figure 1).

The container was placed on the turntable which turns every 90 degree angle.

The NaI detectors measure two discrete gamma-ray energies emitted from the daughter nuclide Pa-234m (1001 keV and 766 keV) of U-238. The measurement of two gamma-rays was designed to correct the matrix attenuation losses.

Three NaI detectors were placed in a line vertically. The one surface of container was divided into 12 regions like as meshes (3 x 4).

The measurement was performed for four sides of a container (12 x 4).

The count time was determined by the need to obtain reasonable count statistics in the two gamma-ray peaks of interest (1001 keV and 766 keV). Normally 900 sec count was adequate. The total measuring time, using 16 scans for each point is four hours.

The weight of each container was registered. These data were then used to determine “activity / unit waste mass” values for defined waste components from which estimates of the total uranium content of filled containers has been established.

The data acquisition system contains a PC-based MCA system, appropriate high voltage power supplies and amplifiers.

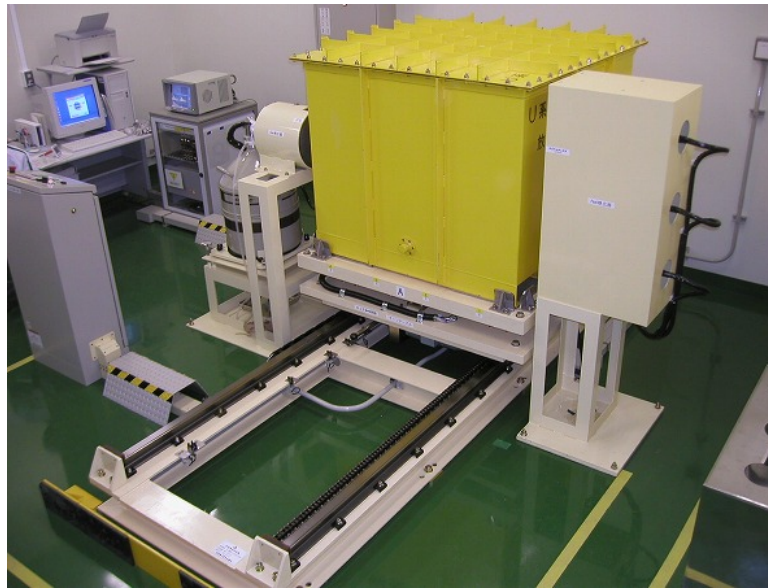


Fig. 1. System configuration

Methods

The measurement was performed on the container divided in imaginary meshes because the distributions of density and radionuclides were not uniform.

We developed a new technique to remove the influence of distance between radionuclide and detector. This technique was named “the Facing Couple Method (FCM)”.

These measurement results confirmed that the quantity of uranium in a large-sized container could be evaluated by passive gamma method (FCM).

The direct radiations emitted by radionuclide in the same direction went through the materials that have the same density. Therefore the effective attenuation distance (ρt) could be evaluated by peak counting rate ratio of two direct radiations. This effective attenuation distance between radionuclide and detector revealed the quantity of uranium in container. In addition, the following relation could be used by the data which is measured at opposing point.

Density of direction of detector (a) may not be the same as direction of detector (c) in the waste, but the distance ($l+2\Delta$) between detector (a) and the detector(c) is constant shown in Figure 2.

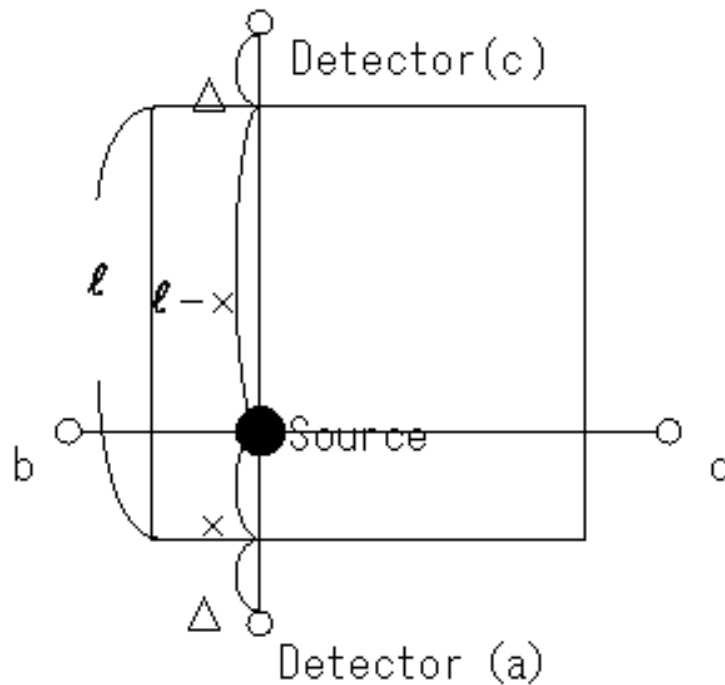


Fig. 2. A conception diagram of FCM

Distance between Source and Detector(a)	$x + \Delta$	(Eq.1)
Distance between Source and Detector(c)	$l - x + \Delta$	
Addition	$l + 2\Delta$	

If reliable measurement data at point (a) and (c) (or (b) and (d)) are provided, the quantity of radionuclide can be estimated except the influence of distance. The estimate process is given below:

At the Detector (a)

$$N_a(1) = A \cdot k_1(1) \cdot k_2(1) \frac{1}{4\pi(x + \Delta)^2} e^{-(\mu_1 \cdot \rho_a \cdot t_a + \mu_{1(Fe)} \cdot \rho_{Fe} \Delta t)} \quad (\text{Eq. 2})$$

2)

$$N_a(2) = A \cdot k_1(2) \cdot k_2(2) \frac{1}{4\pi(x + \Delta)^2} e^{-(\mu_2 \cdot \rho_a \cdot t_a + \mu_{2(Fe)} \cdot \rho_{Fe} \Delta t)}$$

(Eq.3)

Where :

- Na(1) = Counting rate at point (a) of 766 keV.
- Na(2) = Counting rate at point (a) of 1001 keV.
- A = Activity amount of U-238(g).
- k₁(i) = The number of gamma ray / g(U-238) x sec.
- k₂(i) = Counting rate / gamma flux.
- μ = Matrix mass attenuation coefficient (cm² / g).
- ρ = Density of container (g / cm³).
- t = Transmission distance (cm).

$$\frac{N_a(1)}{N_a(2)} = \frac{k_1(1) \cdot k_2(1)}{k_1(2) \cdot k_2(2)} e^{-(\mu_1 - \mu_2) \rho_a t_a - (\mu_{1(Fe)} - \mu_{2(Fe)}) \rho_{Fe} \Delta t}$$

(Eq.4)

Therefore, effective attenuation distance (pt) of gamma ray is decided from peak ratio of counting rate of observation location.

f_a and f_c are defined with Na(2) and Nc(2) as follows:

$$f_a = N_a(2) \cdot \frac{4\pi}{k_1(2) \cdot k_2(2)} \cdot e^{\mu_2 \cdot \rho_a \cdot t_a + \mu_{2(Fe)} \cdot \rho_{Fe} \Delta t} = \frac{A}{(x + \Delta)^2} \quad (\text{Eq.5})$$

$$f_c = N_c(2) \cdot \frac{4\pi}{k_1(2) \cdot k_2(2)} \cdot e^{\mu_2 \cdot \rho_c \cdot t_c + \mu_{2(Fe)} \cdot \rho_{Fe} \Delta t} = \frac{A}{(\ell - x + \Delta)^2} \quad (\text{Eq.6})$$

Finally, x is erased from Eq.5 and Eq.6, and the activity amount (A) is decided as follows.

$$A = (\ell + 2\Delta)^2 \cdot \frac{f_a \cdot f_c}{f_a + f_c + 2\sqrt{f_a \cdot f_c}} \quad (\text{Eq.7})$$

For the equations mentioned above, FCM does not need information of location of source for estimation of activity amount.

Examination

The test was performed to investigate whether the attenuation effect that depends on the distribution of density and the kinds of nuclides was handled in FCM appropriately.

The natural uranium was used as radioactive source.

We analyzed assay results in the following combinations of different patterns of simulated materials.

- A) The place of source in a container: 24points
- B) The quantity of uranium: 50, 100, 500, 1000, 10000 g.
- C) The 10 uranium sources of 100g were placed on the average.
- D) The radioactive source of 100 or 500 g with Fe (1-4.9cm) or PVC tube (1-4.5cm) as shield.
- E) The case of homogeneous system with density 1.0 g/cm^3 that consists of water.
- F) The case of system with density 1.5 g/cm^3 that consists of water, sand and steel. (The container is modeled as having the matrix of the same density as the actual waste one).
- G) Repeatability test.

RESULTS

The result of the relative values which are the observed activity amounts divided by true ones is as follows shown in Figure 3. The relative values in case A fell within 0.45 - 2.08.

The relative ones in case B to D are distributed over 0.61-1.66.

The content in Case F is near to a true waste. In this case, relation ones were distributed over 0.47-1.85. An average value of the relative ones in this test is 0.97.

The standard deviation (σ) was 0.35. In addition, the standard deviation in log-normal distribution is the logarithms of 1.43.

The relationship between weight of waste and detection limit is shown in Figure 4. The relationship is exponential.

The detection limit is 20g when radiation source placed at the center in the inside mesh of a container, at a matrix density of 1 g/cm³.

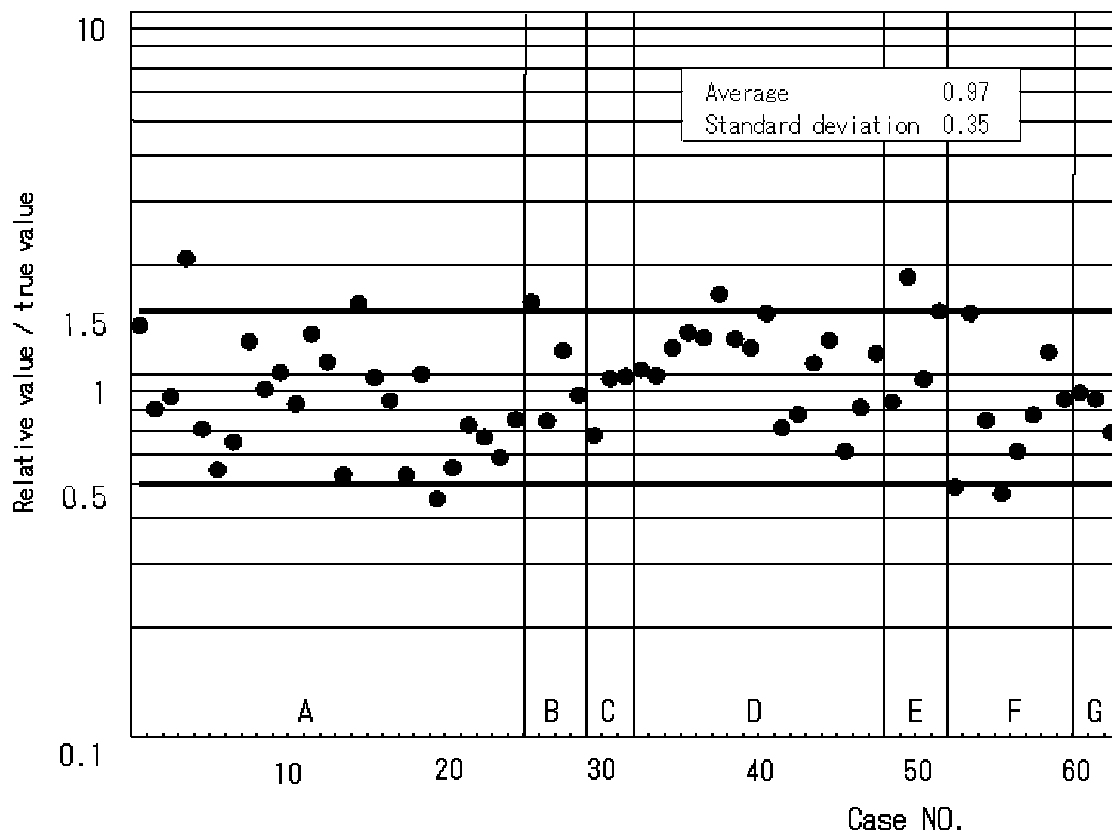
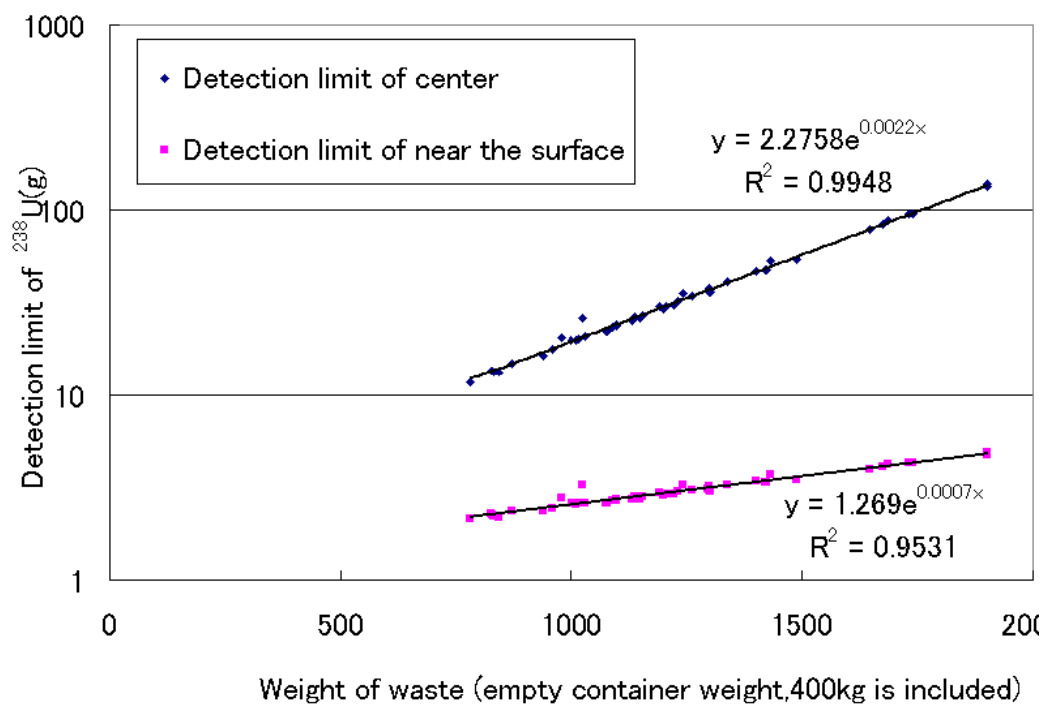


Fig. 3. Result of examination of each case.



Discussion **Fig. 4. Relationship between detection limit and weight of waste**

In this examination the dimensions of a container are approximately 120 x 120 x 90 cm and the variation in the actual distances which gamma rays must travel to escape the matrix is large.

The test with natural uranium clearly shows that the attenuation correction is applied appropriately.

Another test was performed using drums which homogenous contents with uniform distribution of the daughter nuclides of uranium.

We confirmed in this test that the accuracy of results based on the FCM method was equal with results of the conventional homogeneous drum assay system on the passive gamma method.

These facts showed that adequate accuracy was provided by segmented scanning even in a container.

The observation of container started from January 14, 2004 and 107 observations have been completed until August 23, 2004.

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