Development of Estimation Method for Radioactivity Concentration Distribution of Radioactive Wastes in a Box-shaped Container – 17034

Yasushi Nagumo *, Takahiro Tadokoro *, Yuichiro Ueno *, Toshihisa Tsukiyama **, Hideki Yamai**, Takashi Kitahara**
* Hitachi, Ltd. Research & Development Group
** Hitachi-Ge Nuclear Energy, Ltd.

ABSTRACT

We are developing a new estimation method suitable for determining radioactivity concentration distribution of radioactive wastes stored in a square box-shaped container. This method uses the Maximum A Posteriori (MAP) method which is a way to solve the inverse problem, and it estimates the radioactivity concentration distribution by comparing simulated data with measured data. In the measurement, gamma rays emitted from the radioactive wastes are measured by several gamma ray spectrometers which are set at different positions in relation to each other. On the other hand, the gamma ray spectrum data of each spectrometer are simulated by a Monte Carlo simulation code. The method uses an iterative calculation to solve the concentration distribution with measured spectrum data and simulated spectrum data.

We conducted a simulation test to check the algorithm of our estimation method. And we also conducted an experiment using activated objects. We found that the concentration of the wastes in a box-shaped container could be estimated within a 20 % error for the simulation, and within a 40 % error for the experiment and we confirmed that feasibility of our method was favorable.

INTRODUCTION

After the accident at the Fukushima Daiichi Nuclear Power Plant (NPP), as a general rule, the Japanese government set the operating period of NPPs in Japan to 40 years. In this situation, many NPPs that have reached the 40-year limit are expected to be decommissioned. Therefore, rational processing and discarding procedures for radioactive wastes are required.

Drums are conventionally used as containers for low-level radioactive wastes in Japan. Radioactivity concentration of the wastes stored in the drums is measured by using a non-destructive assay (NDA) method. In the NDA method, gamma rays emitted from the radioactive wastes are measured by a germanium radiation detector outside a drum as the latter is being rotated, and the radioactivity concentration is determined based on the measured gamma spectrum data [1]. This method can reduce the effect of localization of radioactivity by measuring the rotating drum, and thus, the concentration of the radioactive wastes is measured accurately.

Recently, in Japan, consideration is being given to square box-shaped containers

instead drums to process, transport, and dispose of the wastes reasonably. If these box-shaped containers are used, the NDA method for measuring cylinder-shaped drums will not be applicable. In that case, an assumption will be made that radioactivity concentration of the wastes in the box-shaped containers is uniform to simplify the determination. However, there is a possibility that the concentration of the radioactive wastes is estimated in excess when there is a concentration distribution or a localization of radioactivity such as a hot spot. Therefore, the method that is able to evaluate radioactivity concentration for the wastes stored in a box-shaped container is required even if there are distributions or localizations.

Then we are developing a new estimation method suitable for determining radioactivity concentration distribution of radioactive wastes stored in a square box-shaped container. In this paper, first of all, the proposed method is explained. After that, the validation test we conducted and the results are described.

PROPOSED ESTIMATION METHOD

The proposed method is one way to solve inverse problems from measured data, and it estimates the radioactivity concentration distribution by comparing measured data with simulated data calculated by a Monte Carlo (MC) simulation code.

Fig. 1 shows the conceptual image of this method. Considering that there is radioactivity concentration distribution, we virtually divide the inner region of the container into several sub regions. In this method, gamma rays emitted from the radioactive wastes are measured by several gamma ray spectrometers which are set at different positions relative to each other. In the MC simulation, the energy spectra of the gamma rays emitted from each sub region are calculated at each detector position. The calculated spectra are registered into a database. These measured spectra and the calculated spectra are used to determine the radioactivity concentration distribution. The flow chart of the procedures in the estimation method is shown in Fig. 2.

From these data, this method estimates a radioactivity concentration distribution by applying the Maximum A Posteriori (MAP) method which is one of the iterative image reconstruction methods used in medical imaging diagnostic apparatuses, such as Positron Emission Tomography (PET) or Single Photon Emission Computed Tomography (SPECT) apparatuses [2]. Although PET and SPECT use counts or count rates of photo peaks emitted from radio isotopes such as F-18, Tc-99m and so on, it is the feature of our proposed method that information about the shape of the gamma ray energy spectrum is used to estimate the distribution under the condition with limited measurement data.

The calculation procedure is shown as below. For comparison with the measured data, first the calculated energy spectrum of detector n, $S_{calc\,n}(E_m)$, is calculated by Eq. 1:

$$S_{calc\,n}(E_m) = \sum_i a_i S_{i\,n}(E_m) \quad (i = 1, \cdots, I), (m = 1, \cdots, M), (n = 1, \cdots, N) \qquad \text{(Eq. 1)}$$

where *i* is the index of a sub region, *I* is the number of sub regions, *m* is the index of an energy bin of the energy spectrum, *M* is the number of energy bins, *n* is the index of a detector, *N* is the number of detectors, *E* is the gamma ray energy, s_{in} is the energy spectrum of detector *n* to detect gamma rays when sub region *i* is the source position, and a_i is the weighting factor applied to the energy spectrum of sub region *i* which is the source position.

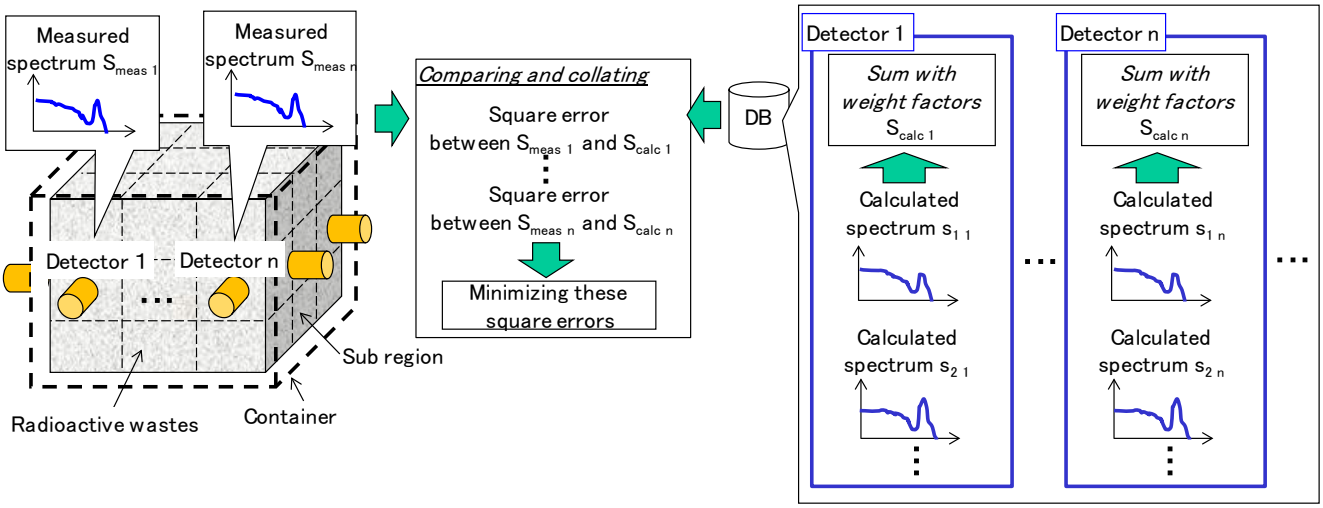


Fig. 1 Conceptual image of the proposed estimation method

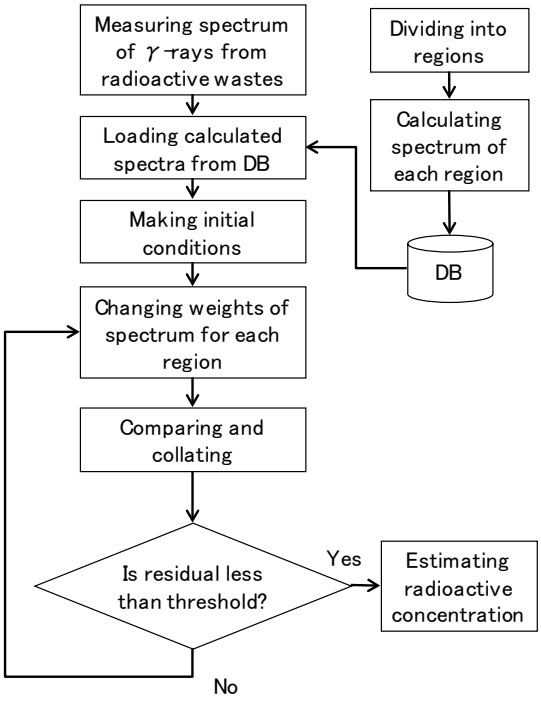


Fig. 2 Flow chart of the proposed estimation method

In this calculation, by updating all of the weighting factor a_i values, the energy spectrum of detector n, $S_{calc\ n}(E_m)$, is iteratively calculated, and $S_{calc\ n}(E_m)$ is brought closer to the measured one, $S_{meas\ n}(E_m)$. The weighting factor a_i is updated by calculating Eq. 2 to Eq. 4 based on the MAP method:

$$P(a_i = w_l) = \frac{B(a_i = w_l)}{\sum_{w_l \in \Omega} B(a_i = w_l)} \quad \text{(Eq. 2)}$$

$$B(a_i = w_l) = \exp\left(-U(a_1, \cdots, a_i = w_l, \cdots, a_I)/T\right) \quad \text{(Eq. 3)}$$

$$\begin{aligned} U(a_1, \cdots, a_i, \cdots a_I) &= \sum_n \sum_m \left(S_{calc\ n}(E_m) - S_{meas\ n}(E_m)\right)^2 \\ &= \sum_n \sum_m \left(\sum_i a_i s_{i\ n}(E_m) - S_{meas\ n}(E_m)\right)^2 \end{aligned} \quad \text{(Eq. 4)}$$

where, $P(a_i = w_l)$ is the probability of transition that the value of the weighting factor a_i is replaced with another one w_i, Ω is a set of available values the weighting factor, T is an arbitrary positive constant value, and $U(a_1,\ldots,a_i,\ldots,a_I)$ is the square error between the calculated energy spectrum and the measured one.

When U is getting smaller, the calculated energy spectrum is brought closer to the measured one and the probability P is getting a higher value. Therefore, the weighting factor a_i giving the highest probability is selected as the optimal factor. This calculation is conducted until convergence. If there is any classification process in which the concentration range of the wastes in the container is gotten before measuring the wastes in a box-shaped container as a screening step, the concentration range of wastes stored in the container can be used as a constrained condition of the MAP method, and the concentration distribution can be estimated more accurately.

VALIDATION TESTING AND RESULTS

Simulation Test

To confirm validity of the algorithm of the proposed method, we conducted a simulation test using a small container geometry. Fig. 3 shows the container geometry for the simulation. The container size was 200mm X 200mm X 200mm, its wall thickness was 10mm, and the container material was steel. The container was filled with concrete materials which were assumed to be radioactive wastes or with concrete materials which were assumed to be filling the container holding stored metal wastes. The number of sub regions in the container was 27 (3 X 3 X 3). The number of detectors was 2 and they were on an outside wall of the box as shown in Fig. 3.

Co-60 was assumed as the measured radionuclide because it is an indicator of a radiated material. Therefore, gamma ray energies of 1.17 MeV and 1.33 MeV of Co-60 were set as emission sources in the MC simulation. The MC simulation code we used in this test was PHITS (Particle and Heavy Ion Transport Code System) [3].

In this simulation test, we gave only one sub region (acting as a hot spot) an 8 times higher concentration than the other sub regions to confirm the basic performance of the proposed estimation method. Measurement data used in this test were simulated with the PHITS MC simulation code. The initial condition of concentration in the container was set to a uniform concentration. Simulations both with and without the constrained condition were also conducted. In the constrained condition, the concentration range was two orders of magnitude.

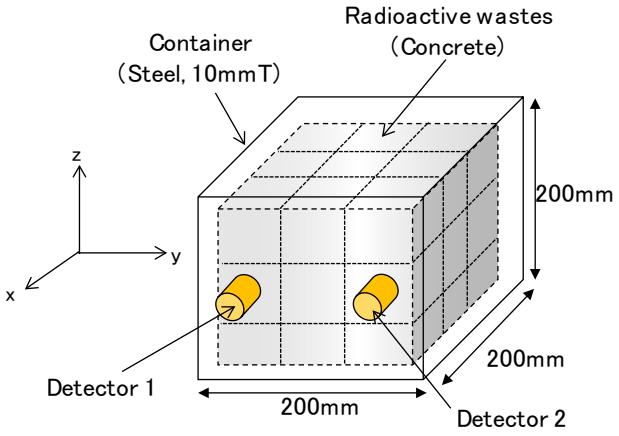
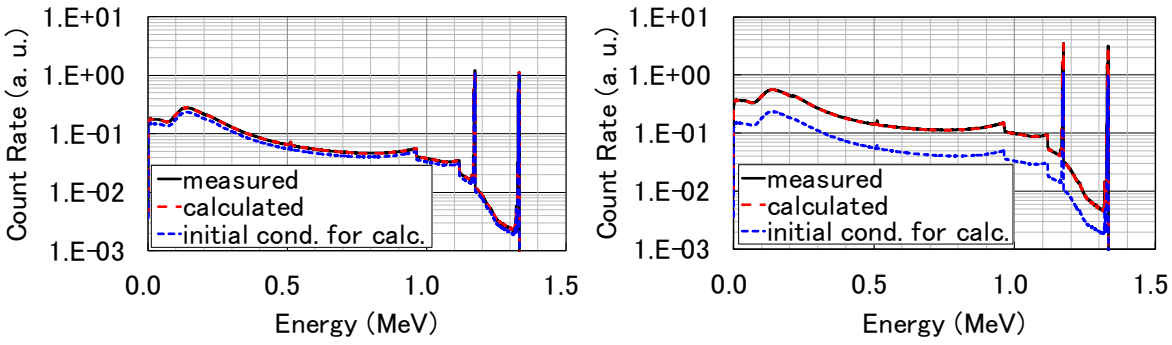
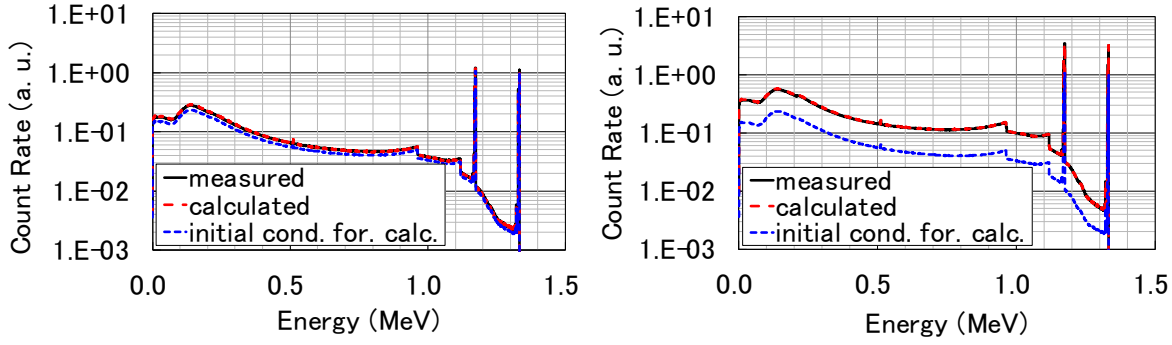


Fig. 3 Container geometry for the simulation

Figs. 4 (a) and (b) are the simulated spectra for the two detectors with the constrained condition and without it, respectively. We confirmed that simulated spectra of both detectors accorded well with the measured spectra for the two cases. Fig. 5 shows the correct radioactivity concentration distribution and the simulation radioactivity concentration distributions for the cases with and without the constrained condition. In the case without the constrained condition, the results showed that only 2 sub regions had any radioactivity concentration and other sub regions had none. On the other hand, in the case with the constrained condition, the results were better and the obtained concentration distribution was closer to the correct distribution. The results of the radioactivity concentration of the whole container in the case with the constrained condition are shown in Fig. 6. The horizontal axis indicates distance from the point at the center between detectors 1 and 2 to the center of the hot spot-containing sub region. The vertical axis represents the concentration of the whole container. We confirmed that the concentration was within a ± 20 % error regardless of the hot spot position.



(a) With constrained condition



(b) Without constrained condition

Fig. 4 Simulated spectra (left, Detector 1; right Detector 2; as indicated in Fig. 3)

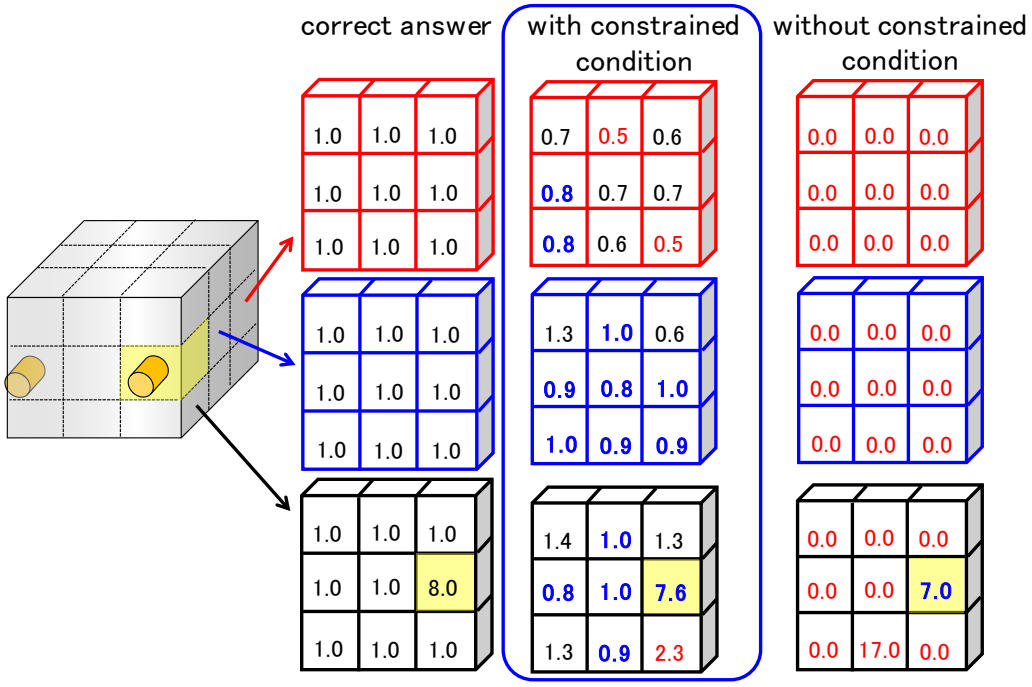


Fig. 5 Simulated radioactivity concentration distributions

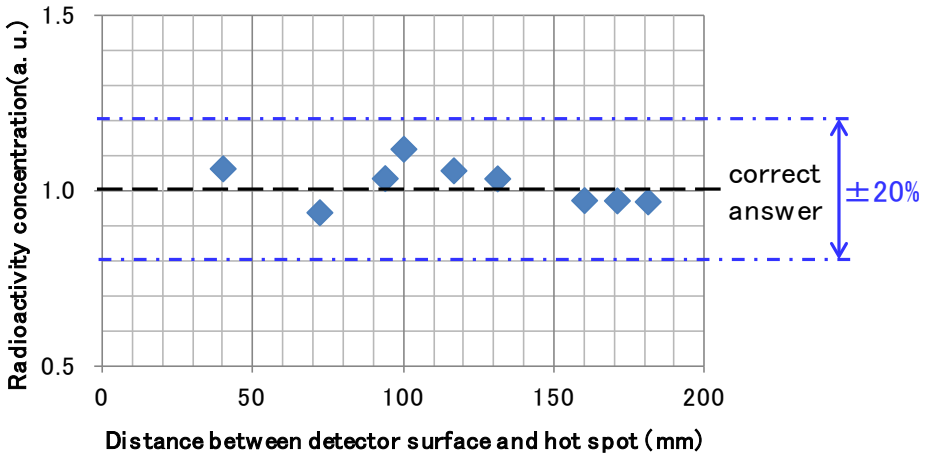


Fig. 6 Concentration results of the whole container with the constrained condition

Performance Test

To confirm basic performance of the proposed estimation method, we carried out a performance test.

Fig. 7 shows equipment for the test which consisted of a box-shaped container having the same size and being made of the same material as the containers that are expected to be used in the decommissioning work. The container was 1.3m X 1.3m X 1.3m and made of steel. We used the GR1-A® gamma spectrometer (Kromek). A positioning tool system to move and set the spectrometer at the measuring positions was also used.

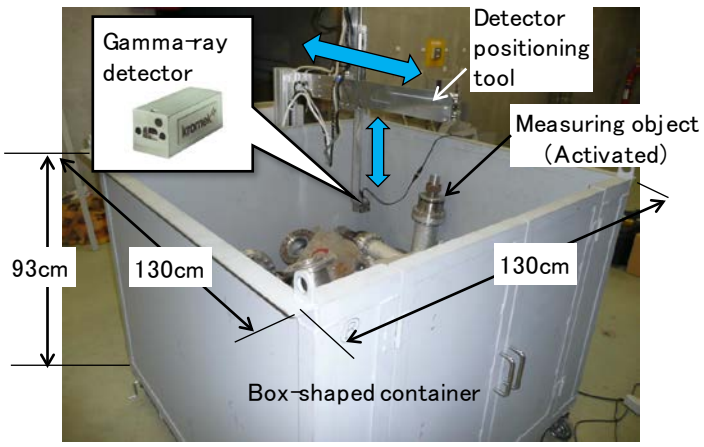


Fig. 7 Equipment for the performance test

Fig. 8 shows the objects we used in this test. They were parts from an actual

accelerator that had been used and so were activated, with gamma rays of Co-60 mainly being emitted. Total weight of these objects was about 445 kg. This weight was used in the MC simulation.

We supposed an actual storage state of the wastes, meaning that the objects were placed in the container in a disorderly fashion as shown in Fig. 8. The index numbers of the divided regions, virtual division lines, and measuring positions are also indicated in the figure. In the performance test, the inner region of the container was virtually divided into 3 X 3 sub regions. The height of all measurement positions was 50 cm from the bottom of the container to avoid interference between the spectrometer and the objects. The measurement time at each position was 30 minutes. The constrained condition that we applied set the concentration range as two orders of magnitude including the measurement results obtained with the germanium (Ge) detector, described as follows.

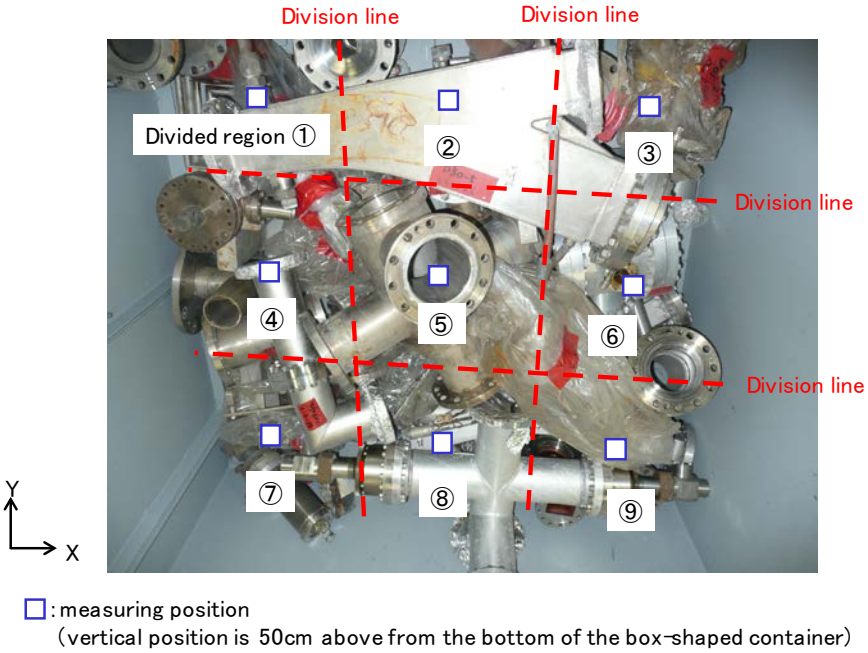
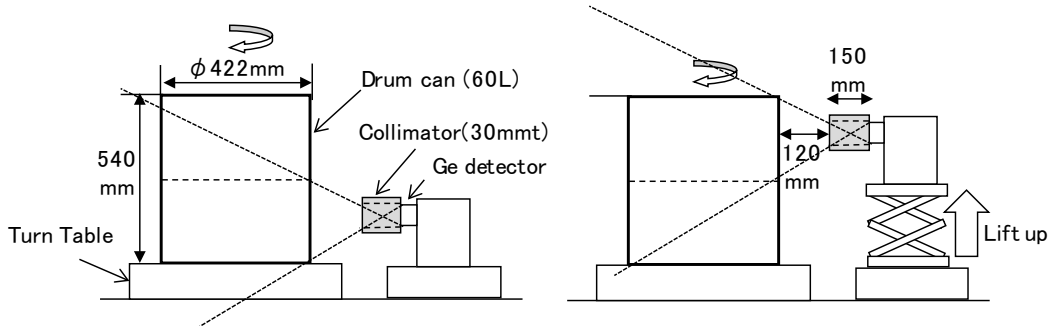


Fig. 8 Objects used in the performance test and their random placement as viewed from the top of the container

The amount of radioactivity of the objects to compare with the results of our proposed method was obtained by measuring them with a Ge detector. Fig. 9 shows schematic images of the measurement with the Ge detector. These objects were split into 9 groups based on the sub regions as described above. Then, the objects of each group were placed into the drum and measurements were made with the Ge detector which had a cylindrical lead collimator attached to it while the drum was turning. The top half and the bottom half of the drum were measured separately, and the amount of radioactivity was obtained by comparing these measured data with the results of PHITS simulation for the drum.



(a) Measuring the drum lower part (b) Measuring the drum upper part

Fig. 9 Schematic images of the measurement with Ge detector for the drum

Fig. 10 compares the estimated spectrum of the detector of divided region No.5 with the measured spectrum as an example. We confirmed that the estimated spectrum accorded well with the measured spectrum. Fig. 11 compares the estimated radioactivity concentration distribution with data measured by the Ge detector. The estimated concentration distribution qualitatively agreed with the data measured by the Ge detector. The concentration results of the whole container are also shown in Fig. 11. The error of the estimation was 39.98 %, so we confirmed that the proposed method was able to determine the radioactivity concentration within a 40% error.

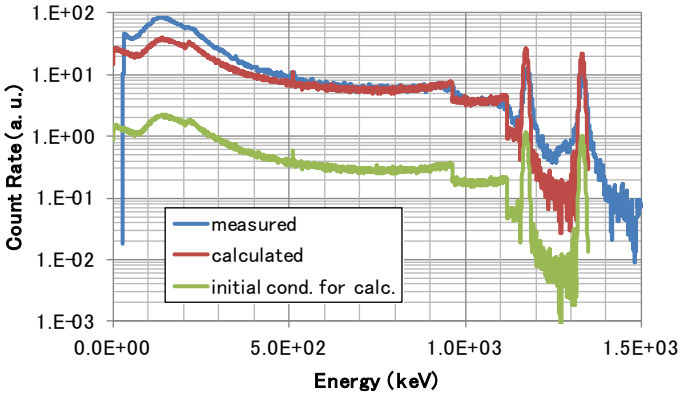


Fig. 10 An example of the estimated and the calculated spectra (divided region No. 5)

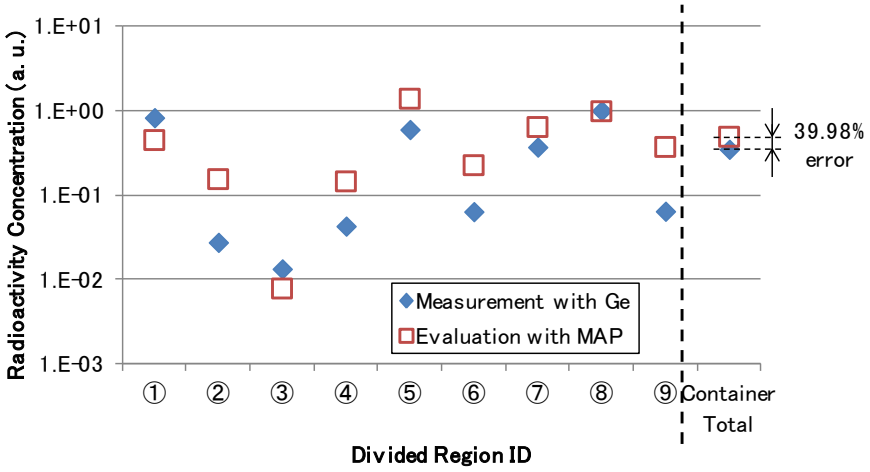


Fig. 11 Estimated radioactivity concentration distribution compared with data measured by Ge detector

CONCLUSIONS

We are developing a new estimation method for evaluating radioactivity concentration distribution of radioactive wastes stored in a box-shaped container. This method is based on the MAP method and it estimates the radioactivity concentration distribution by comparing simulated data with measured data of individual detectors.

We conducted a simulation test to check the algorithm of the estimation method and we did a performance test using activated objects. We found that the concentration of the wastes in a box-shaped container could be estimated within a 20 % error for the simulation, and within a 40 % error for the experiment. We confirmed that application of our method was feasible. In the future, we are going to improve the method so that it estimates the radioactivity concentration distribution more accurately.

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2. H. Kudo and H. Nakamura, "Attenuation Map Reconstruction Using Topology Constrained Labeling," Electronics and Communications in Japan, Vol. 86, No. 4, pp.31-41 (2003)
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