Journal of Physics: Conference Series

PAPER • OPEN ACCESS

In-situ determination of residual specific activity in activated concrete walls of a PET-cyclotron room

To cite this article: H Matsumura *et al* 2018 *J. Phys.: Conf. Ser.* **1046** 012016

IOP Conf. Series: Journal of Physics: Conf. Series **1046** (2018) 012016 doi:10.1088/1742-6596/1046/1/012016

In-situ determination of residual specific activity in activated concrete walls of a PET-cyclotron room

H Matsumura[1], A Toyoda[1], K Masumoto[1], G Yoshida[1], T Yagishita[2], T Nakabayashi[2], H Sasaki[2], K Matsumura[2], Y Yamaya[2] and Y Miyazaki[3]

[1]Radiation Science Center, High Energy Accelerator Research Organization (KEK) 1-1 Oho, Tsukuba, Ibaraki 305-0801, Japan
[2]Japan Environment Research Co., Ltd., 6-24-1 Nishi-Shinjuku, Shinjuku-ku, Tokyo 160-0023, Japan
[3]The Medical and Pharmacological Research Center Foundation, Wo-32, Inoyama-machi, Hakui, Ishikawa 925-0613, Japan

hiroshi.matsumura@kek.jp

Abstract. In the decommissioning work for concrete walls of PET-cyclotron rooms, an in-situ measurement is expected to be useful for obtaining a contour map of the specific activity on the walls without destroying the structure. In this study, specific activities of γ-ray-emitting radionuclides in concrete walls were determined by using an in-situ measurement method employing a portable Ge semiconductor detector, and compared with the specific activity obtained using the sampling measurement method, at the Medical and Pharmacological Research Center Foundation in Hakui, Ishikawa, Japan. Accordingly, the specific activity could be determined by the in-situ determination method. Since there is a clear correlation between the total specific activity of γ-ray-emitting radionuclides and contact dose rate, the specific activity can be determined approximately by contact dose-rate measurement using a NaI scintillation survey meter. The specific activity of each γ-ray-emitting radionuclide can also be estimated from the contact dose rate using a NaI scintillation survey meter. The in-situ measurement method is a powerful tool for the decommissioning of the PET cyclotron room.

1. Introduction

Radioactive isotopes, such as ^{18}F, ^{15}O, ^{13}N, and ^{11}C, are produced as tracers for positron emission tomography (PET). Proton cyclotrons with acceleration energies of between ten and twenty MeV are used to produce these radioactive isotopes. There are more than 140 cyclotrons for medical use in Japan, and it is estimated that many of them will be decommissioned due to aging in the near future. Therefore, the decommissioning methodology for these accelerator facilities is an important issue.

Radioactive isotopes for PET are produced by proton-induced nuclear reactions in production targets. Neutrons are also produced simultaneously in the nuclear reactions. The produced neutrons flight into the accelerator body and the concrete walls of the accelerator room. Consequently, the neutrons induce nuclear reactions in these materials, and activate them. For the decommissioning of the accelerator facility, “radioactive materials” must be isolated. The concrete walls in the accelerator room are especially huge. The establishment of a determination method for residual specific activity in the concrete walls is important for the efficient decommissioning of accelerator facilities.

Before the decommissioning of the accelerator facility, the residual specific activity in the concrete walls has to be determined to isolate the radioactive material region. At present, the determination of specific activity in concrete is carried out by using γ-ray spectrometry with a Ge semiconductor detector (Ge detector) after sampling by boring the concrete core [1, 2]. In this "sampling measurement method", the depth variations of the residual specific activities of radionuclides can be determined.

However, the sampling measurement method has the following two drawbacks. (1) Specific skills for radiological work are required for the concrete sampling. (2) Since the sampling number is limited, it is difficult to map for identifying the whole image of activation. If the residual specific activity in the concrete wall is determined non-destructively by attaching a detector to the concrete wall (in-situ measurement method), a contour map of the specific activity can be obtained in detail without destroying the structure.

The establishment of the in-situ measurement method can improve the efficiency of the decommissioning process and reduce radioactive waste and disposal cost as much as possible. Therefore, in this study, we determined the residual specific activity in concrete walls by using a portable Ge detector and a NaI scintillation survey meter (NaI survey meter) which were brought into an actual PET cyclotron room during decommissioning. The sampling measurement was also carried out, and the validity of the determination results was evaluated by a comparison between the in-situ measurement method and the sampling measurement method.

2. Experimental procedures

2.1. Investigation of a PET-cyclotron facility
An NKK-Oxford superconducting cyclotron [3] used for the production of radiopharmaceuticals for PET at the Medical and Pharmacological Research Center Foundation in Hakui, Ishikawa, Japan was decommissioned in March 2015. The concrete walls of this PET cyclotron room were investigated in this study. Information on the PET-cyclotron facility is given in Table 1.

The energy and output current of the proton beam are 12 MeV and 60 μA, respectively. Since the cyclotron was used for the production of PET radiopharmaceuticals for delivery to hospitals, the operation time per day of the cyclotron (~7 h) was much longer than that of general production use at a hospital (~1 hour or less). Consequently, the activation level in the cyclotron facility was estimated to be eight times higher than that in general hospital cyclotron facilities, considering the operation time of the cyclotron and neutron flux in the room.

The operation time of the PET-cyclotron every year between 1998 and 2014 is shown in figure 1. Although the operation period is from 1998 to 2015, most of the 17618 hours of operation time occupies the ten years from 2005–2014. The decommissioning work began immediately in 2015, and the entire cyclotron body and its accessories were removed from the cyclotron room. Currently, only the concrete walls of the cyclotron room remain unchanged.

Table 1. Information on the PET-cyclotron facility investigated in this study.

Accelerator	Superconducting cyclotron (NKK/Oxford)
Beam	Particle: protons, Energy: 12 MeV, Output current: 60 μA
Purpose of use	Production of PET radiopharmaceuticals for delivery to hospitals
Operation start	July, 2000 (commissioning in 1998 and 1999)
Decommissioning	March, 2015
Integral operation time	17618 h
Room size	5.6 m (north-south) and 5.2 m (east-west)
Height of a ceiling	2.7 m

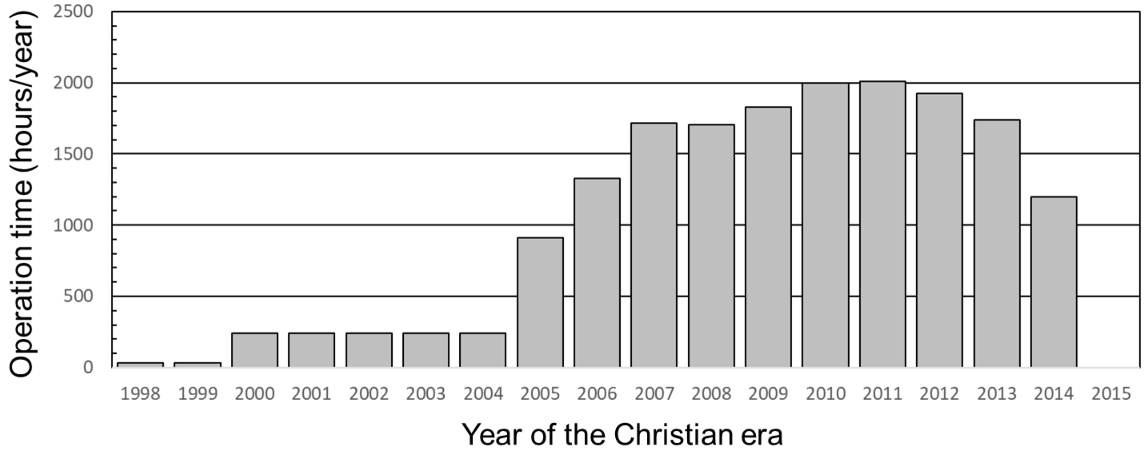

Figure 1. Operation time of the PET-cyclotron each year at the Medical and Pharmacological Research Center Foundation in Hakui, Ishikawa, Japan.

2.2. Sampling measurement method using Ge detector

The specific activities of γ-emitting radionuclides in the concrete walls were determined after concrete core boring. The locations where the concrete core boring was performed are shown as CW1, CW2, CW4, and CW5 in figure 2. The details of this method are described below.

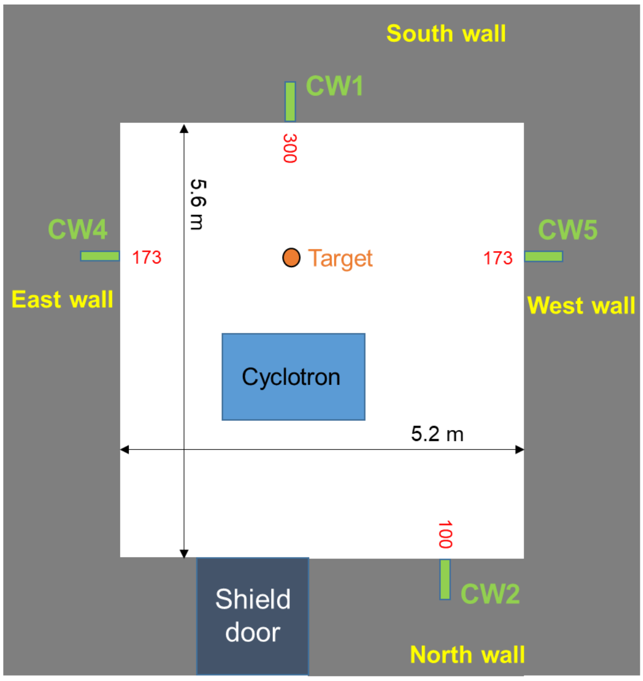

Figure 2. Core boring locations of CW1, CW2, CW4, and CW5. The height of the core boring is 131 cm from the floor. The red values indicate the distances of the CW4 and CW5 cores from the south wall and the distances of the CW1 and CW2 cores from the west wall in cm unit.

First, a 1.5-cm-thick gypsum board was cut and removed from the surface of the concrete walls. Second, 6.5-cm-diameter concrete cores (length: ~50 cm) were collected from the concrete walls. Third, the cores were sliced into 2-cm-thick disks and then milled with a stamp mill. Finally, the powdered concrete samples were transferred into U8 plastic containers and γ-ray spectrometry was performed using Ge detectors to determine the specific activities.

The γ-ray energies used for analysis were 344 keV for ^{152}Eu, 1333 keV for ^{60}Co, 605 keV for ^{134}Cs, 835 keV for ^{54}Mn, and 1461 keV for ^{40}K. The detection efficiencies of the Ge detectors were determined using a standard γ-ray source mixed ^{57}Co, ^{60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{137}Cs, ^{139}Ce, and ^{203}Hg having the same shape as the samples. The nuclear data were taken from [4]. The specific activities of ^{152}Eu, ^{60}Co, ^{134}Cs, and ^{54}Mn in January 2017 could be obtained.

2.3. Specific activity measurement of a fraction of the reinforcing rod

A fraction of the reinforcing rod (diameter: 1.9 cm, length: ~5.5 cm) was found in the CW1 concrete core. The specific activity of ^{60}Co in the isolated reinforcing rod was determined by γ-ray spectrometry using a Ge detector. The Ge detector was characterized for the efficiency calculation software, ISOCS, by Canberra Inc. [5, 6]. The detection efficiencies of a ^{60}Co 1333-keV γ-ray from the fraction of the reinforcing rod were obtained by using ISOCS. The nuclear data were taken from [4].

2.4. In-situ measurement method using a Ge detector

For in-situ measurements, a portable Ge detector was kept in contact with the surface of the concrete walls. The in-situ measurements were performed at twenty locations in January 2017. They are shown in figure 3. To limit the detection to close range from the Ge detector, the detector was shielded from ambient γ rays by a 6.5-cm-thick and 22.0-cm-long cylindrical Pb shield with a hole of 9.0-cm diameter. Measurements of γ rays were typically performed for ~1500 s (1399–57792 s). The γ-ray energies used for the analysis were 1408 keV for ^{152}Eu, 1333 keV for ^{60}Co, 605 keV for ^{134}Cs, 723 keV for ^{154}Eu, 835 keV for ^{54}Mn, and 1461 keV for ^{40}K. The nuclear data were taken from [4].

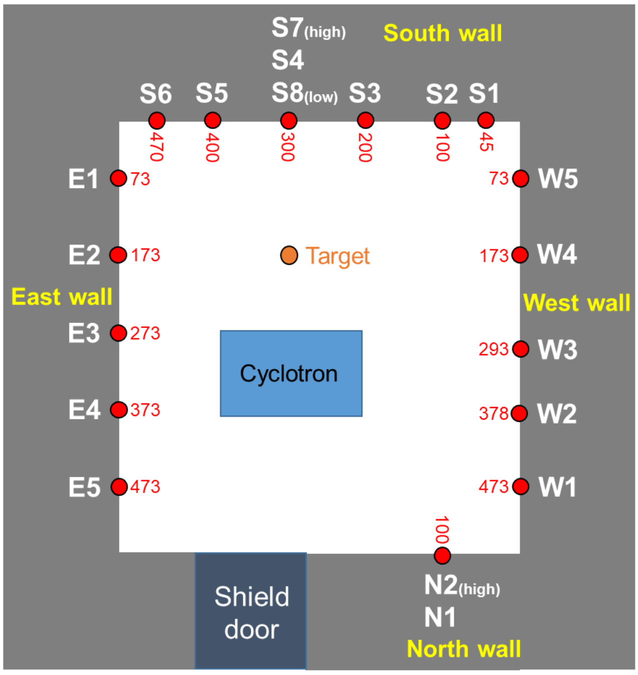

Figure 3. Twenty locations where the in-situ measurement was performed using the Ge detector (red circles). Basically, the height of the measurement was 91 cm from the floor. The heights of S8, S7, and N2 are 51, 151, and 151 cm from the floor, respectively. The red values indicate the distances of the locations of W1-W5 and E1-E5 from the south wall and the distances of the locations of S1-S8, N1, and N2 from the west wall in cm unit.

The portable Ge detector was characterized for ISOCS. The detection efficiency for γ rays from the concrete walls was obtained by using ISOCS, assuming that the γ-ray source was distributed uniformly in the concrete wall. The measured density (2.30 g/cm^3) and analysed chemical composition were input for the efficiency calculation. In the uniform activity condition, the attainable region of γ rays, where the count contribution occupied over 90%, depend on the γ-ray energy. The attainable regions were estimated to have diameter and depth of approximately 40 cm and 17 cm for ^{152}Eu, ^{60}Co, and ^{40}K, respectively, and 30 cm and 13 cm for ^{134}Cs, ^{154}Eu, and ^{54}Mn, respectively, by using ISOCS. The specific activities of ^{152}Eu, ^{60}Co, ^{134}Cs, ^{154}Eu, and ^{54}Mn in the concrete walls were determined.

2.5. *In-situ contact dose rate measurement using a NaI survey meter*

In January 2017 (at the same time when the in-situ measurement using Ge detector was carried out), a NaI survey meter was also attached to the surface of the concrete walls, to obtain a correlation curve between the specific activity and contact dose rate. The measurements of the contact dose rate were performed at the same locations as those of the Ge detector measurements, as shown in figure 3. The model of the NaI survey meter used for this measurement was TCS-171 (Hitachi Aloka Medical, Ltd.). The NaI survey meter was shielded by the same Pb shield as the Ge detector described in Section 2.4. The time constant was set to 10 s. The measurement at a location was performed for a time longer than 30 s. The contact dose rate from a natural background, such as ^{40}K, U series, and Th series in concrete, was obtained as 0.07 μSv/h by measuring a concrete wall in a non-activated adjoining room.

3. Results and discussion

3.1. *Depth profiles of the specific activities*

By the sampling measurement method using Ge detectors described in Section 2.2, the depth profiles of the specific activities of ^{152}Eu, ^{60}Co, ^{134}Cs, and ^{54}Mn in the concrete walls were obtained. The depth profiles of the specific activities from the CW1, CW2, CW4, and CW5 concrete core samples have been plotted in figure 4. The specific activities in decreasing order were ^{40}K, ^{152}Eu, ^{60}Co, ^{134}Cs, and ^{54}Mn. Especially, the specific activities of ^{152}Eu and ^{60}Co account for a substantial fraction of the total specific activity, except for natural ^{40}K. Both ^{152}Eu and ^{60}Co are important radionuclides for the decommissioning of the concrete walls.

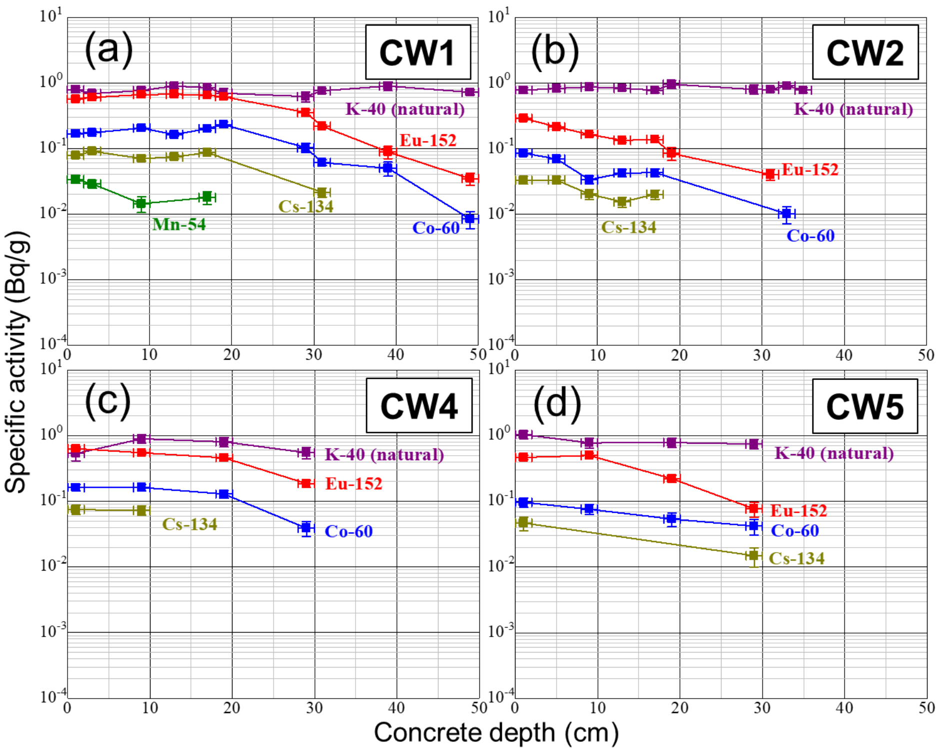

Figure 4. Depth profiles of the specific activities of ^{152}Eu, ^{60}Co, ^{134}Cs, ^{54}Mn, and ^{40}K in the concrete walls from (a) CW1, (b) CW2, (c) CW4, and (d) CW5 concrete core samples in January 2017.

The depth profiles were similar among the radionuclides. Furthermore, the depth profiles were similar among the four different walls. Typically, the specific activity was approximately constant from the surface to a depth of ~20 cm. Thereafter, the specific activity was found to decrease exponentially. In the in-situ measurement, most of the counting comes from the depth shallower than 17 cm, as described in Section 2.4. The attainable region of the γ rays agreed with the constant region of the specific activity. Therefore, the distribution of the γ-ray source in the concrete walls could be considered uniform for the in-situ measurement. This means that the specific activities could be determined approximately by the in-situ measurement method using the Ge detector.

3.2. Spatial distributions of the specific activities

In Section 3.1, it was found that the specific activities could be determined approximately by the in-situ measurement method. Figure 5 shows the spatial distributions of the specific activities of ^{152}Eu, ^{60}Co, ^{134}Cs, ^{154}Eu, ^{54}Mn, and ^{40}K in the concrete walls, which were determined by the in-situ measurement method using the Ge detector, as described in Section 2.4.

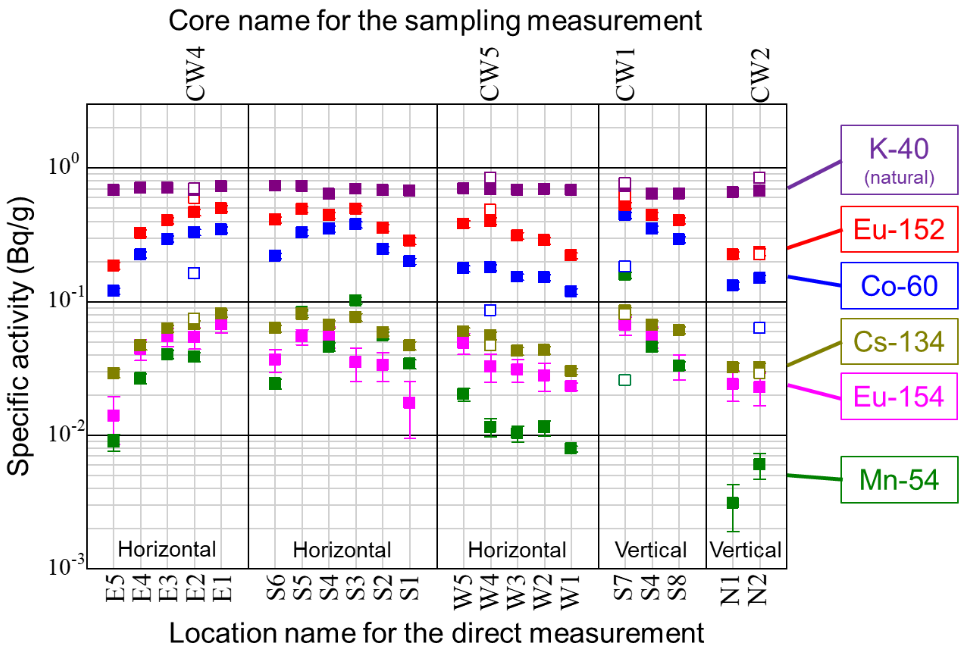

Figure 5. Spatial distributions of the specific activities of ^{152}Eu, ^{60}Co, ^{134}Cs, ^{154}Eu, ^{54}Mn, and ^{40}K in the concrete walls in January 2017. The closed squares indicate the specific activities determined by the in-situ measurement method using the Ge detector. The open squares indicate averages of the specific activities from 0 to 10 cm depth determined by the sampling measurement method.

The distribution of the ^{40}K specific activity was constant because of the inclusion of ^{40}K naturally in the concrete walls. The specific activity distributions were similar among ^{152}Eu, ^{60}Co, ^{134}Cs, and ^{154}Eu. The thermal neutron capture reaction is common in these radionuclide productions. Near the target, the activation level was high. The specific activity decreased with increasing distance from the target. The distribution of the ^{54}Mn specific activity was different from that of the others. In the concrete walls, ^{54}Mn would be produced by ^{54}Fe(n, p)^{54}Mn. Since the specific activity of ^{54}Mn is much smaller than that of the others, ^{54}Mn is not an important radionuclide for the decommissioning of the concrete walls.

In figure 5, the specific activities determined by the in-situ measurement method (closed squares) are compared with those determined by the sampling measurement method (open squares). The specific activities of ^{152}Eu, ^{134}Cs, ^{154}Eu, and ^{40}K determined by the in-situ measurement method agreed well with those obtained by the sampling measurement method.

However, in every concrete wall, the specific activity of ^{60}Co determined by the in-situ measurement method was higher than that in the sampling measurement method. The ratios of the ^{60}Co specific activities (sampling/in-situ) were approximately constant (average: 0.45) regardless of the location. In this facility, 10-cm mesh grids of reinforcing rods with 1.9-cm diameter were placed at a depth of 5 cm from the concrete surface. The reinforcing rods as well as the concrete walls were exposed to thermal neutrons. In the reinforcing rods, ^{60}Co could be produced by the thermal neutron capture reaction of the ^{59}Co impurity. A fraction of the reinforcing rod was obtained from the CW1 concrete core, as described in Section 2.3. The specific activity of ^{60}Co in the isolated reinforcing rods was determined as 3.04±0.15 Bq/g in January 2017.

Next, the count rate of 1333-keV γ-rays from the ^{60}Co of reinforcing rods in the in-situ measurement method was estimated using the measured specific activity in the reinforcing rod and counting efficiency calculated by ISOCS. Accordingly, the count rate ratio of the ^{60}Co γ rays ([estimated from the reinforcing rod]/[measured]) was estimated to be 0.58, which corresponded to the ratios of 0.42 of the ^{60}Co specific activities ([sampling measurement]/[in-situ measurement]). This value agreed well with the measured ratio of 0.45. Therefore, the discrepancy in the ^{60}Co specific activity in the concrete walls between the sampling measurement method and in-situ measurement method was attributed to the contribution from the reinforcing rods. In the in-situ measurement method, the ^{60}Co specific activities in the concrete walls were obtained by multiplying 0.45 as a correction factor.

3.3. Specific activity ratios of each radionuclide for ^{152}Eu

The specific activity ratios of each radionuclide for ^{152}Eu in the concrete walls determined by the in-situ measurement method are plotted in figure 6. The specific activity ratios were independent of the location (except for ^{54}Mn). This fact indicated that if the specific activity of ^{152}Eu is known in some way, the specific activities of the others could be estimated by using the specific activity ratios. The specific activity ratios were determined as 0.24 for ^{60}Co/^{152}Eu, 0.15 for ^{134}Cs/^{152}Eu, 0.105 for ^{154}Eu/^{152}Eu, and 0.096 for ^{54}Mn/^{152}Eu.

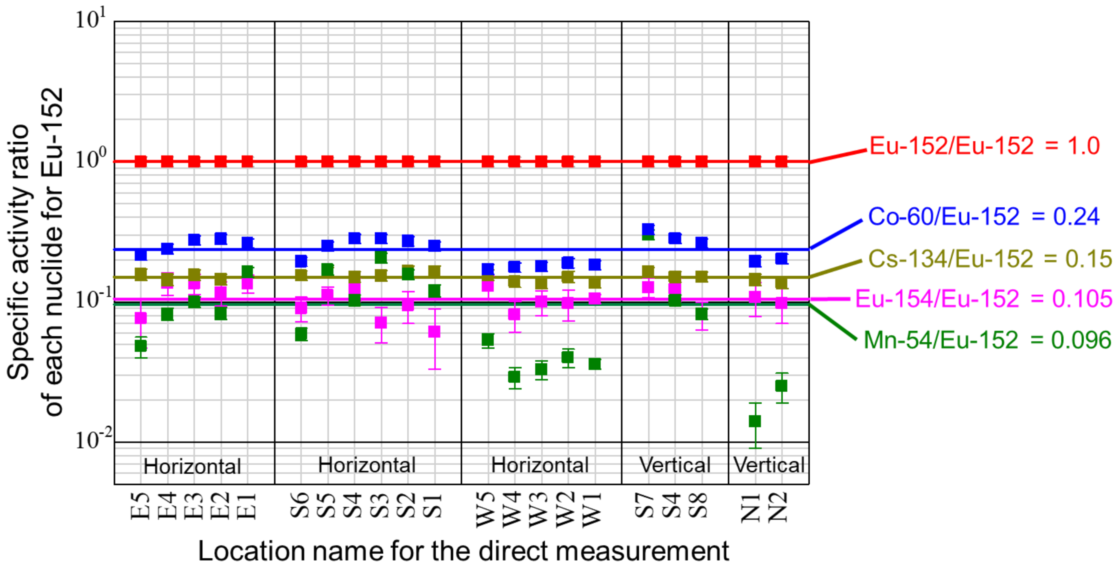

Figure 6. Specific activity ratios of each radionuclide for ^{152}Eu in the concrete walls. The lines indicate the averages of the ratios. The ^{60}Co specific activities corrected for the interference from the reinforcing rods were used for calculating the specific activity ratios of ^{60}Co/^{152}Eu.

3.4. Correlation between the total specific activity and net contact dose rate

The in-situ measurements of contact dose rates were carried out using the NaI survey meter as described in Section 2.5. A correlation curve between the net contact dose rate and the total specific activity in the concrete walls is shown in figure 7. The net contact dose rate means a dose rate obtained by subtracting the natural background described in Section 2.5. The total specific activity was the sum of the specific activities of ^{152}Eu, ^{60}Co, ^{134}Cs, ^{154}Eu, and ^{54}Mn, which were determined by the in-situ measurement method using the Ge detector. There is a clear correlation between the total specific activity and net contact dose rate. Therefore, the net contact dose rate on the concrete wall (D) can be used for estimating the total specific activity in the concrete wall (A_{total}), as A_{total} (Bq/g) = D (μSv/h) ×2.1.

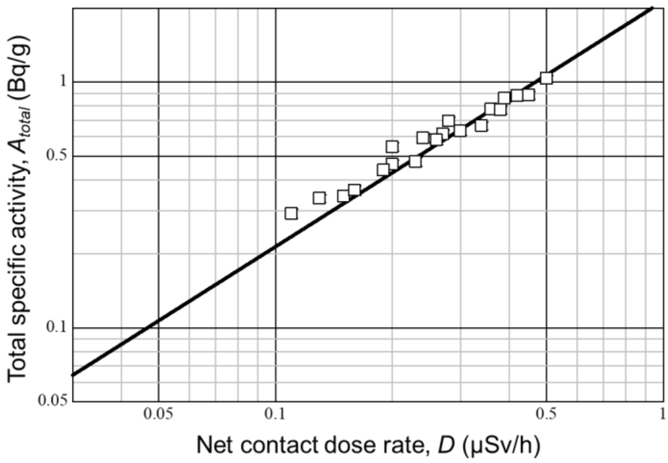

Figure 7. Correlation curve between the net contact dose rate and the total specific activity in the concrete walls.

3.5. Estimation of specific activities of the radionuclides from the net contact dose rates

A correlation curve between the net contact dose rate and specific activity of ^{152}Eu in the concrete walls is shown in Figure 8. There is also a clear correlation between the ^{152}Eu specific activity (A_{Eu-152}) and the net contact dose rate (D), as A_{Eu-152} (Bq/g) = D (μSv/h) ×1.3.

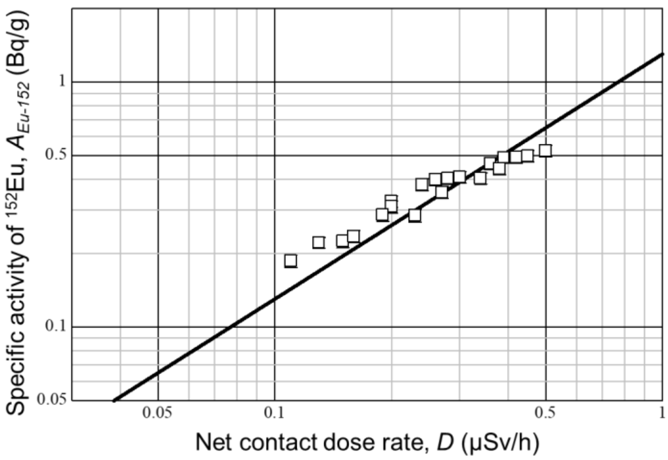

Figure 8. Correlation curve between the net contact dose rate and specific activity of ^{152}Eu in the concrete walls.

In Section 3.3, it was found that the specific activity ratios of each radionuclide for ^{152}Eu in the concrete walls were approximately constant, regardless of the location. Therefore, the specific activities of ^{60}Co, ^{134}Cs, ^{154}Eu, and ^{54}Mn can be estimated from the net contact dose rate and the specific activity ratio of each radionuclide for ^{152}Eu. In Figure 9, the specific activities estimated from the net contact dose rate and the specific activity ratio of each radionuclide for ^{152}Eu (open circles) are compared with the in-situ measurement method using the Ge detector (closed squares). Both the specific activities were in good agreement (except for ^{54}Mn). Thus, the net contact dose rate is available for estimating the specific activity of every radionuclide.

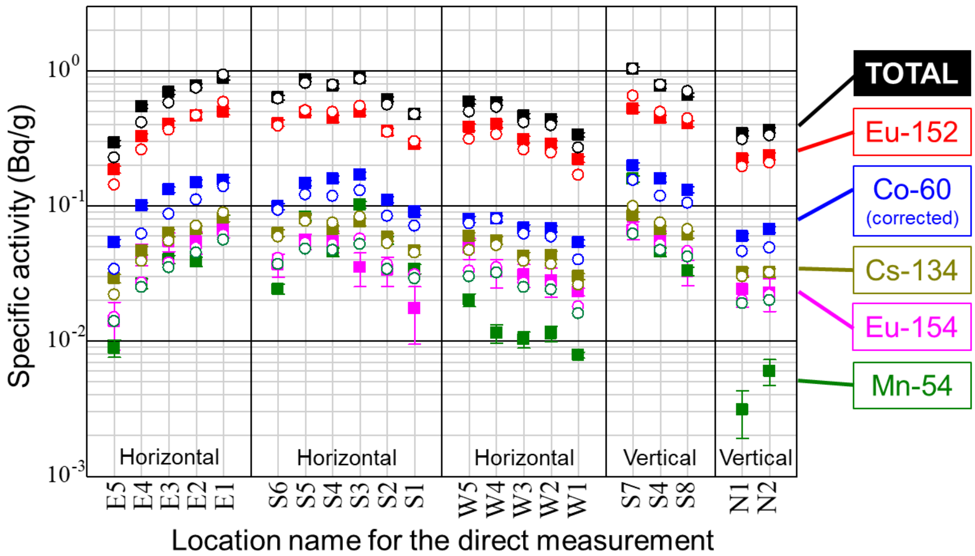

Figure 9. Comparison of the specific activities of ^{152}Eu, ^{60}Co (corrected), ^{134}Cs, ^{154}Eu, ^{54}Mn, and their sums in the concrete walls between by the in-situ measurement method using the Ge detector (closed squares) and by estimation from the net contact dose rates measured using the NaI survey meter (open circles).

4. Conclusions

The dominant radionuclides after ~2 years from the end of operation were found to be ^{152}Eu and ^{60}Co for the decommissioning of the concrete walls of a PET cyclotron room at the Medical and Pharmacological Research Center Foundation in Hakui, Ishikawa, Japan. Since the depth profile of specific activity was uniform approximately from the surface to a depth of ~20 cm, the specific activity could be determined by the in-situ determination method using a portable Ge detector.

There is a clear correlation between the total specific activity of γ-ray-emitting radionuclides and net contact dose rate. Therefore, the specific activity can be determined approximately by contact dose-rate measurement using a NaI survey meter. The specific activity ratios of ^{60}Co, ^{134}Cs, and ^{154}Eu for ^{152}Eu were independent of the location. The specific activity of each γ-ray-emitting radionuclide can also be determined approximately by contact dose-rate measurement using a NaI survey meter.

The in-situ measurement method is a powerful tool for the decommissioning of a PET cyclotron room because a contour map of the specific activity can be obtained in detail without destroying the structure.

References

[1] Masumoto K, Matsumura H, Bessho K and Toyoda A 2008 *J. Radioanal. Nucl. Chem.* **278** 449

[2] Wang Q B, Masumoto K, Bessho K, Matsumura H, Miura T and Shibata T 2004 *J. Radioanal. Nucl. Chem.* **262** 587
[3] Kusumoto K, Mizuno K and Iwata R 1992 *Proceedings of the IVth International Workshop on Targetry and Target Chemisty (TARGETRY'91), September 9-12, 1991, PSI Villigen, Switzerland, in PSI-Proc.* **92-01**
[4] Firestone R B 1996 *Table of Isotopes,* 8th ed., John Wiley and Sons, Inc., New York
[5] Venkataraman R, Bronson F, Atrashkevich V, Field M and Young B M 2005 *J. Radioanal. Nucl. Chem.* **264** 213
[6] Bronson F L 2003 *J. Radioanal. Nucl. Chem.* **255** 137