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Depth Profiles of Radionuclides Induced in Shielding Concrete of the 12 GeV Proton Accelerator Facility at KEK

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The depth profiles of long-lived radionuclides were measured for the shielding concrete exposed to secondary particles in the 12 GeV proton beam-line tunnel at KEK. Various long-lived radionuclides, ^{7}Be, ^{22}Na, ^{46}Sc, ^{51}Cr, ^{54}Mn, ^{56}Co, ^{57}Co, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{134}Cs, ^{152}Eu and ^{154}Eu, were observed in concrete samples by γ-ray spectra measurements, and ^{3}H was also detected by liquid scintillation counting. The major radionuclides induced in the ordinary concrete were ^{3}H, ^{7}Be and ^{22}Na and ^{3}H, ^{54}Mn and ^{60}Co in heavy concrete, respectively. It was confirmed that the maximum concentration of the radionuclides induced by thermal neutron capture reactions, such as ^{46}Sc, ^{51}Cr, ^{60}Co, ^{65}Zn, ^{152}Eu and ^{154}En, etc., was located at about 20 cm in depth from the surface of shielding concrete. The fast neutron attenuation lengths for ordinary and heavy concrete were estimated to be 131 g/cm^2 and 150 g/cm^2 from depth profiles of ^{54}Mn, respectively. These values were in agreement with the values determined using activation detectors at KEK.

KEYWORD: depth profile, radionuclide, shield concrete, high-energy accelerator, spallation reaction, thermal neutron capture, attenuation length, neutron flux, fast neutron, heavy concrete, ordinary concrete

I. Introduction

The 12 GeV proton synchrotron of the High Energy Accelerator Research Organization (KEK) has been utilized for high-energy physics and its related experiments for about twenty years since the first experiment started. The shielding concrete of the slow extracted proton beam line (EP2 beam line) of this facility has been exposed to high-energy primary and/or secondary particles during machine operation. As a result, a variety of radionuclides, including tritium, have been produced in the shielding concrete through both nuclear spallation reactions and thermal neutron capture reactions of concrete elements.

The activated shielding concrete is one of the major sources of radiation exposure to radiation workers, such as operating crew and maintenance workers, and become the most important problem of radioactive waste management in the reconstruction of high-energy accelerators. Thus quantitative information about radionuclides induced in the shielding concrete is very important not only for evaluating radiation doses for radiation workers, but also for planning the reconstruction or decommissioning of an accelerator facility. Several measurements of residual radioactivities in the concrete wall of an operating accelerator have been reported(1)(2)(3). There were, however, few studies on a high-energy proton accelerator.

In this study, the depth profiles of various long-lived radionuclides induced in the shielding concrete of the EP2 beam line were determined using γ-ray spectroscopy and a liquid scintillation counting method. Then, the neutron fluence rate in the EP2 beam line and fast neutron attenuation length in concrete were estimated.

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II. Sample Preparation and Measurement

A plane view of the EP2 beam line and 4 sampling positions are shown in **Fig. 1**. Positions 1 and 3 are on the concrete wall and positions 2 and 4 are on the concrete floor (ordinary concrete). The measurement samples were cored out of the shielding concrete (3~4 m in length for the concrete wall and 40~60 cm for the concrete floor with diameter of 5 cm) at the end of the long summer shutdown (1st and 2nd October) which had started on 17th June, so as to keep the radiation exposure for workers at a low level.

There are three beam lines, 1.7 m high from the floor level, and three production targets in each beam line for the production of secondary particles, such as K mesons and π mesons, etc. The beam was accelerated to 12 GeV and the average intensity was 2×10^{12} ppp during operation. **Fig. 2** shows a cross section view of position 1, which is the horizontal beam line, and just beside the production target (Pt: 6 mm $\phi \times$ 4 cm) in which 30 % of the primary beam are lost. The shielding wall, 1.5 m far from the target and 4 m in thickness, consists of four kinds of shielding blocks: each shielding block is 1 m thick and from the inside pyrite concrete, ordinary concrete, iron block and magnetite concrete. Position 2 is located on the concrete floor just below the EP2-C line and about 1.7 m upstream from the target. Position 3 is located on the side shield, 1.5 m from the beam duct and at the same level. The shielding wall consisted of two kinds of shielding concrete: from the inside, ordinary concrete 1 m and magnetite concrete 2 m. Position 4 was located on the concrete floor just below the EP2-B line. The line loss of the beams was 2×10^{-4}/m.

Samples taken at several points of cored concrete bars, whose constituents are listed in **Table 1**, were grained into powder of

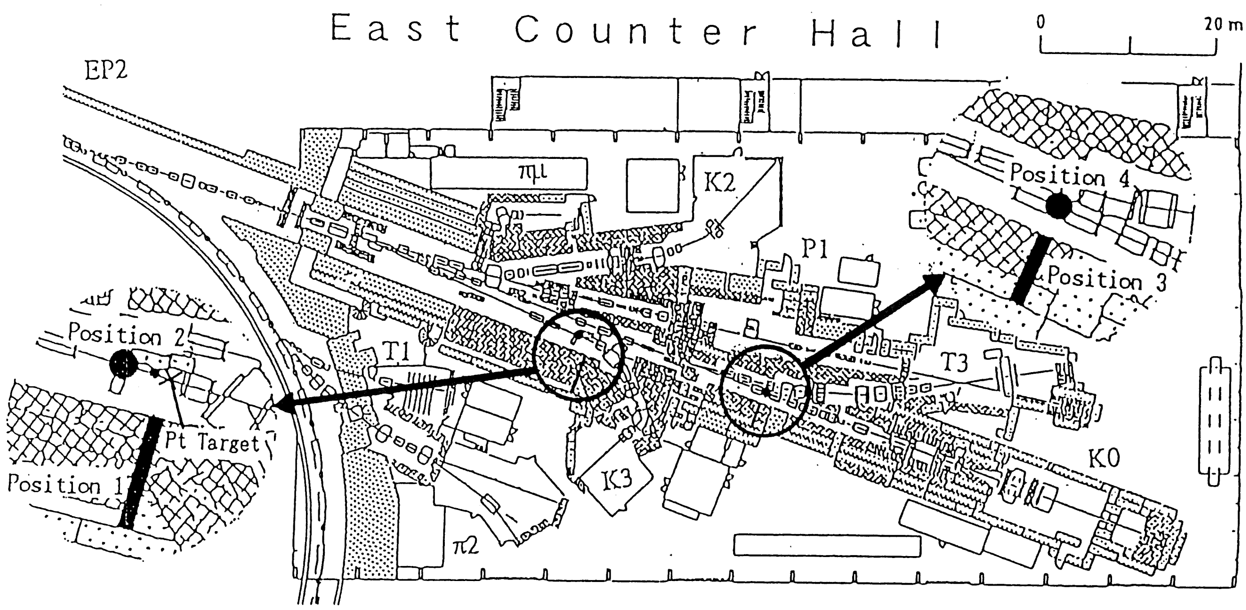

Fig. 1 A plane view of EP2 beam line and 4 sampling positions

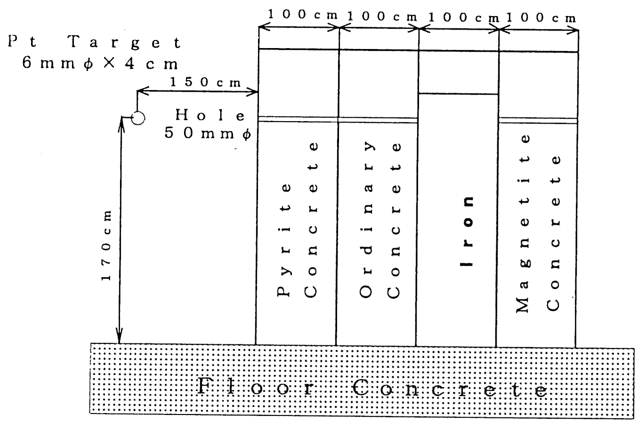

Fig.2 A cross section of position 1

Table 1 The results of elementary analysis of concrete constituents by ICP

	Na (%)	Al (%)	Mn (ppm)	Fe (%)	Co (ppm)
Position 1					
Pyrite	0.62	1.33	2000	49.3	36
Ordinary	1.67	5.65	400	2.08	<6
Magnetite	0.26	0.9	110	33.2	176
Position 2					
Ordinary	1.56	6.52	570	2.43	<6
Position 3					
Ordinary	1.81	8.83	480	1.73	<6
Magnetite	0.31	0.79	140	34.5	100
Position 4					
Ordinary	1.64	6.28	500	2.32	<6

100 μm. The radioactivities induced in the concrete samples were measured with a Pure-Ge detector system for gamma-emitters and liquid scintillation counter for ^{3}H. From the results of radioactivity measurements, the depth profiles of various radionuclides in the shielding concrete were obtained and the average neutron flux was estimated.

III. Results and Discussion

1. Depth Profiles

The radioactivities of ^{7}Be, ^{22}Na, ^{46}Sc, ^{51}Cr, ^{54}Mn, ^{56}Co, ^{57}Co, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{134}Cs, ^{152}Eu and ^{154}Eu induced in the concrete samples were determined from the γ–ray measurements and ^{3}H from liquid scintillation counting, respectively. In these radionuclides, ^{46}Sc, ^{51}Cr, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{134}Cs, ^{152}Eu and ^{154}Eu ware created by a thermal neutron capture reaction. On the other hand, ^{3}H, ^{7}Be, ^{22}Na, ^{54}Mn, ^{56}Co and ^{57}Co were produced by nuclear spallation reactions of fast neutrons. The nuclear data of the main radionuclides are listed in **Table 2**.

The most abundant radionuclides were ^{3}H in ordinary concrete and ^{54}Mn in heavy (pyrites and magnetite) concrete. Major long-lived radionuclides induced in ordinary concrete were ^{3}H, ^{7}Be, ^{22}Na, ^{46}Sc, ^{54}Mn, ^{60}Co, ^{134}Cs and ^{152}Eu and ^{3}H, ^{22}Na, ^{46}Sc, ^{54}Mn, ^{60}Co, ^{65}Zn and ^{152}Eu in heavy concrete, respectively. The shielding wall at position 1 (heavy concrete) near to the Pt target was of course strongly activated and the total concentration of radionuclides was about 830 Bq/g. **Figures 3** and **4** show depth profiles of the principal long-lived radionuclides induced in the concrete samples at position 1 and 3, respectively.

The depth profiles depend slightly on the type of nuclear reactions. The maximum concentrations of ^{60}Co and ^{152}Eu induced in the concrete by thermal neutron capture reactions were observed at about 20 cm in depth from the inside surface of the shielding concrete. On the other hand, the concentration of radionuclides produced by spallation reactions of fast neutrons decreased according to the increase in the depth from

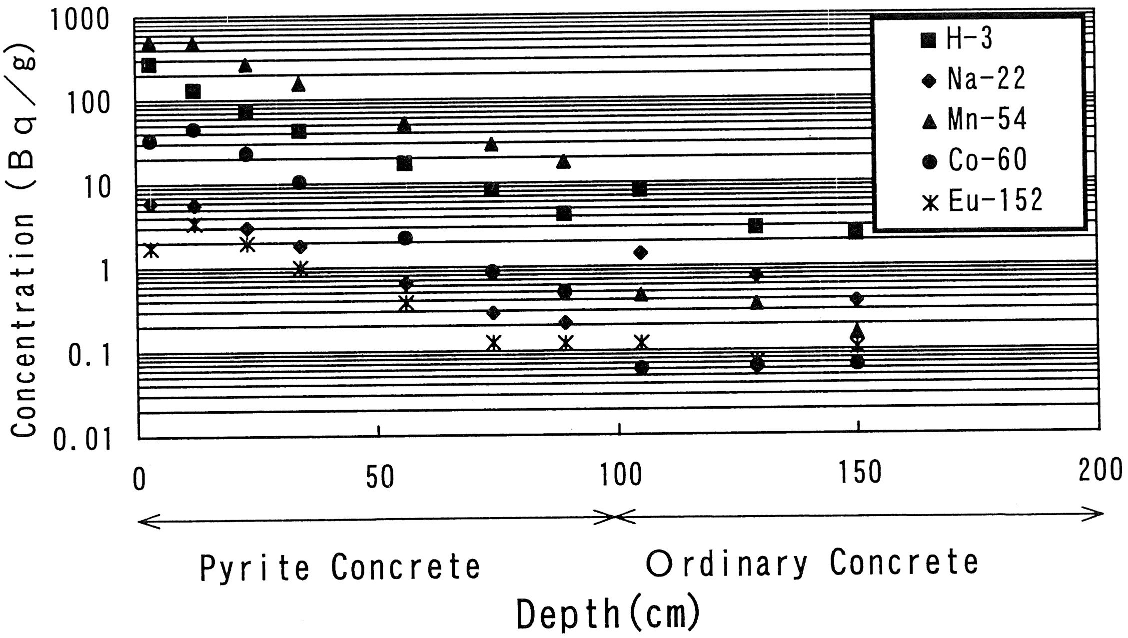

Fig. 3 Depth profiles of principal radionuclides induced in the concrete samples at position 1.

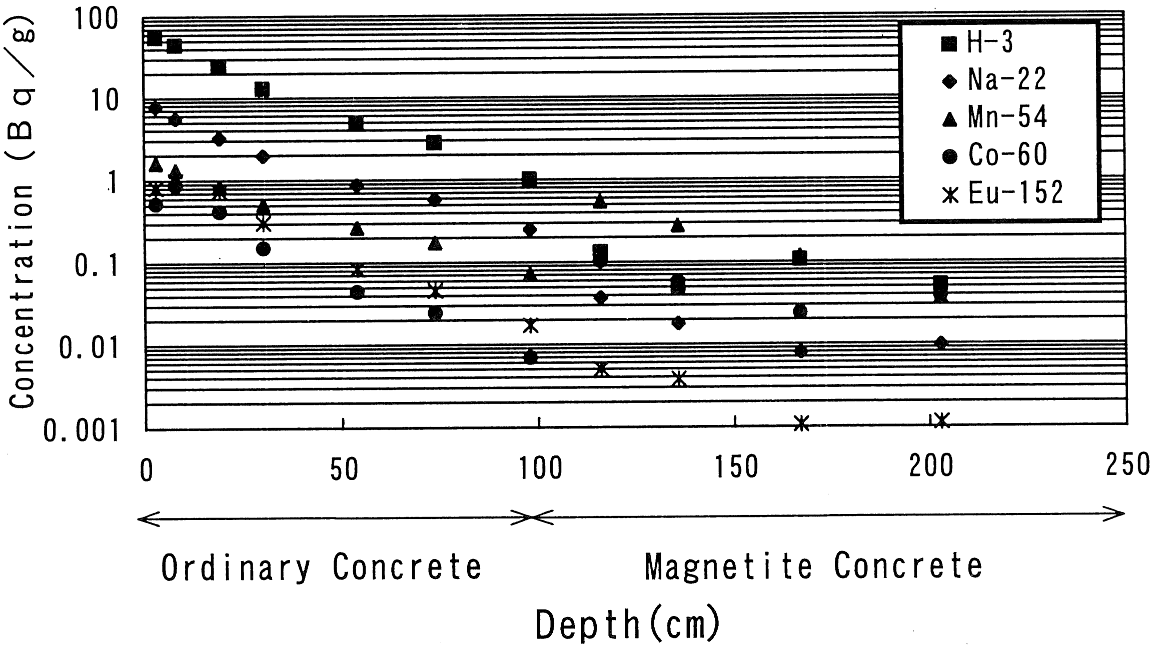

Fig. 4 Depth profiles of principal radionuclides induced in the concrete samples at position 3.

the inside surface of the shielding concrete.

2. Neutron Fluence Rate

Assuming that the redioactivities of ^{54}Mn and ^{60}Co were saturated and that ^{60}Co was created exclusively by thermal neutrons, the fast and thermal neutron fluence rates were estimated from ^{54}Mn and ^{60}Co radioactivities based on the Fe and Co concentrations of the shielding concrete samples at positions 1 and 3. The thermal neutron fluence rates were estimated to be 2.4 X 10^6 n/cm^2/s at position 1 and 2.3 X 10^5 n/cm^2/s at position 3, respectively, whereas the fast neutron fluence rates were 2.7 X 10^6 n/cm^2/s at position 1 and 2.5 X 10^5 n/cm^2/s at position 3. Both the thermal and fast neutron fluence rates depended on the location. The neutron fluence rate of the shielding concrete surface near to the target is higher than that of beam line by about one order. **Figure 5** shows the depth profiles of ^{54}Mn and ^{60}Co induced in the concrete samples at position 2 and 4. Those results give the same information.

3. Fast Neutron Attenuation Length

The fast neutron fluence rate ϕ_p from the target can be expressed in the 90° direction to the beam as follows:

$$\phi_p = A \times \exp(-d/\lambda_p)/r^2, \qquad (1)$$

Table 2 Nuclear data of main radionuclides induced in concrete

Nuclide	Half Life	Reaction	Cross section(barn)	Abundance (%)
Na-22	2.6y	(n, 2n)	0.017 (14.6MeV)	100
Sc-46	83.8d	(n, γ)	26.5	100
Mn-54	313.d	(n, p)	0.61	5.84
Co-60	5.27y	(n, γ)	37	100
Eu-152	13.3y	(n, γ)	5900	47.7

where r is the distance from the target, d is the thickness of the shield and λ_p is the attenuation length. And, of course the radioactivity induced in the shielding concrete is proportional to neutron fluence rate. Then, according to eq(1), the fast neutron attenuation lengths (λ_p) in heavy concrete were estimated to be 94 g/cm^2 and 150 g/cm^2 from depth profiles of ^{22}Na and ^{54}Mn in Fig. 3, respectively.

Also, at the location in which a line source is dominant, the fast neutron fluence rate ϕ_l can be expressed by the following approximate equation:

$$\phi_l = B \times \exp(-d/\lambda_l)/r \qquad (2).$$

From the ^{22}Na and ^{54}Mn depth profiles in Fig. 4, the fast neutron attenuation lengths (λ_l) in ordinary concrete were estimated to be 95 g/cm^2 and 131 g/cm^2, respectively.

The attenuation lengths of fast neutron estimated from ^{54}Mn depth profiles are in agreement with 143 g/cm^2 in ordinary concrete and 163 g/cm^2 in heavy concrete within the statistical error, measured by Ban et al.(4) using activation detectors at KEK. But the values estimated from ^{22}Na depth profiles were slightly smaller than those values.

IV. Conclusion

In order to obtain the quantitative information about the radioactivity induced in shielding concrete, the depth profiles of long-lived radionuclides induced in the shielding concrete

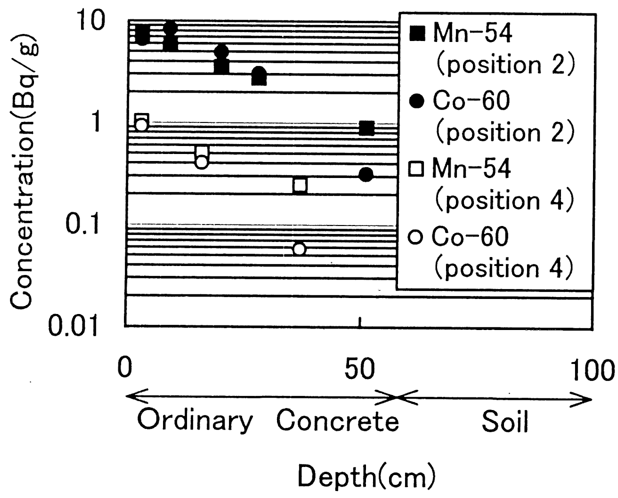

Fig. 5 Depth profiles of ^{54}Mn and ^{60}Co induced in the concrete samples at positions 2 and 4.

of the beam-line tunnel of the 12 GeV proton accelerator facility at KEK were determined using γ-ray and β-ray counting methods. ^{3}H, ^{7}Be, ^{22}Na, ^{46}Sc, ^{51}Cr, ^{54}Mn, ^{56}Co, ^{57}Co, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{134}Cs, ^{152}Eu and ^{154}Eu were observed in the concrete samples. Major radionuclides induced in the shielding concrete were ^{3}H, ^{22}Na, ^{54}Mn, ^{60}Co and ^{152}Eu. It was confirmed that the radionuclides induced by the thermal neutron capture reactions showed the highest concentrations at about 20 cm in depth from the inside surface of the shielding concrete. The fast neutron attenuation lengths estimated from the depth profile of ^{54}Mn in ordinary and heavy concrete were in agreement with the value determined using the activation detectors at KEK.

— References —

(1) Phillips, A.B., *et al.* : *Health Phys.*, **51**, 337 (1986).
(2) Urabe, I., *et al.* : *Health Phys.*, **60**, 587 (1991).
(3) Kimura, K., *et al.* : *Health Phys.*, **67**, 621 (1994).
(4) Ban, S., *et al.* : *Nucl. Instrum. Methods*, 174, 271 (1980).