MEASUREMENT AND ANALYSIS OF INDUCED ACTIVITIES IN CONCRETE IRRADIATED USING HIGH-ENERGY NEUTRONS AT KENS NEUTRON SPALLATION SOURCE FACILITY

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Precise estimation of induced activities in concrete shields for high-energy accelerator facilities is one of the most important issues that need to be solved, not only for the reduction of exposure for workers, but also for the reduction of radioactive wastes. Irradiation experiments have been performed by using the 500 MeV Neutron Spallation Source Facility in KEK. The large concrete assembly was placed in the direction of 0° to the beamline. Two kinds of samples were placed at several positions in the assembly. The irradiation period was about 1 week and induced activities in the samples were measured until ~1.5 y after irradiation. From the comparison between the experiment and the available Monte Carlo calculation code system, good agreement was obtained for 24Na, 47Sc, 47Ca and 54Mn within a factor 2; however, large discrepancies were observed for some other nuclides.

INTRODUCTION

Constructions of high-energy proton and heavy-ion accelerator facilities, such as Radio Isotope Beam Factory (RIBF)(1) of The Institute Of Physical And Chemical Research (RIKEN) and Japan Proton Accelerator Research Complex (J-PARC)(2) of Japan Atomic Energy Research Institute (JAERI) and High Energy Accelerator Research Organization (KEK), have just started. From the safety point of view, not only radiation shielding but also precise estimation of induced activities of construction materials, such as concrete, is one of the most important issues during maintenance periods for exposure to workers and after shutdown for radioactive wastes.

As far as concrete is concerned, there exists no concrete which chemical compositions are exactly the same as each other. The experimental result of only one kind of concrete could not estimate all other kinds of concretes. In the case of estimation of shielding ability of concrete, the principle component of concrete is very important, but for the estimation of radioactive wastes, the very rare components, such as cobalt whose composition varies from 0.1 to 100 ppm, play an important role.

However, fortunately the components of several concretes, such as O, Si, Al, Fe, Ca, Na and K, were same, although their compositions were different. The best way to estimate the induced activities in each concrete practically is by using the calculation code system verified by the typical concrete activation experiment.

In this study, benchmark experiments have been performed by using typical ordinary concrete samples. The objects of this study are to acquire the characteristics of induced activities in concrete, to verify the available calculation code system and to clarify the issues that need to be solved to improve the accuracy of the calculation code system for high-energy accelerator facilities.

MATERIALS AND METHODS

Experimental layout

Irradiation experiments have been performed at the KENS high-energy beam course of KEK. The experimental layout, vertical and horizontal cross-sectional view, is shown in Figure 1. High-energy neutrons were produced in the tungsten target, bombarded with 500 MeV protons. The concrete assembly, whose dimensions were 3 m width, 2.1 m height and 4 m depth, was placed in the existing ordinary concrete wall. The distance between the target and the front surface of the assembly was 2.5 m. Seven slots in the assembly were prepared in the direction of 0° to the beamline for the samples of the irradiation experiment.

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Samples and irradiation conditions

The samples of the irradiation experiment were of two kinds of concrete. One was an ordinary concrete whose composition was equivalent to the concrete assembly. The other one was a limestone aggregate that was considered low-activation concrete. The dimensions of the sample were 60 mm diameter and 10 mm thickness and were sealed in acrylic cases of 1 mm thickness. Irradiation periods were $\sim 7$ d and average beam current at the target was estimated from 4.7 to 5.8 mA.

Measurement

The induced activities in the samples were measured using germanium detectors. The standard sources of the same shape and same density were prepared for the determination of the detector efficiency, to reduce the uncertainty of efficiency correction, such as self shielding by the samples and sum peak effects.

EXPERIMENTAL RESULTS

From the measurement after shutdown to 1 y, 37 radioactive nuclides, $^{44K}$, $^{34Cl}$, $^{38Cl}$, $^{49Cr}$, $^{39Cl}$, $^{41Ar}$, $^{56Mn}$, $^{43Sc}$, $^{44Sc}$, $^{52Fe}$, $^{42K}$, $^{24Na}$, $^{55Co}$, $^{28Mg}$, $^{48Cr}$, $^{43K}$, $^{48Sc}$, $^{44Sc}$, $^{87Y}$, $^{45Sc}$, $^{47Ca}$, $^{52Mn}$, $^{56Ni}$, $^{131Ba}$, $^{48V}$, $^{51Cr}$, $^{7Be}$, $^{148Eu}$, $^{58Co}$, $^{56Co}$, $^{46Sc}$, $^{57Co}$, $^{54Mn}$, $^{134Cs}$, $^{60Na}$, $^{60Co}$ and $^{152Eu}$, were observed in the order of half-life.
Figures 2 and 3 show the experimental results of some radioactive nuclides for two kinds of concrete samples, which were obtained in the wide range of depth of the concrete assembly. The induced activities (Bq g\(^{-1}\)) were saturated activities for the comparison of induced activities between the ordinary concrete and the limestone one. The cooling period was ~2 d. The horizontal axis is the distance from the surface of the assembly, and the vertical one is the induced activities.

The induced activities of these two samples changed exponentially except at the front surface of the assembly. This fact indicated that the shape of the neutron spectra in the concrete assembly is almost the same. The induced activities of \(^{24}\text{Na}\), \(^{52}\text{Mn}\) and \(^{54}\text{Mn}\) in limestone are much smaller than those in the ordinary concrete.

Figure 4 shows the induced activities of long-lived radioactive nuclides of the ordinary concrete samples. The cooling period was ~1 y. The induced activities were normalised at the end of irradiation. They also decreased exponentially as the depth of concrete assembly increased.

**CALCULATION**

Figure 5 shows the block diagram of the calculation code system. The neutron spectra emitted from the tungsten target bombarded by high energy protons in the direction of 0-degree to the beam line were calculated by using the NMTC/JAM calculation code. Induced activities in the samples produced by high-energy neutrons, \(E_n > 20\ \text{MeV}\), were calculated by using the NMTC/JAM calculation code. Yield data obtained by using the NMCT/JAM calculation code were input for the DCHAIN-SP2001\(^{4)}\) to calculate the induced activities at any cooling period by using decay data for each activity. Radioactivities induced in the samples by lower energy neutrons, \(E_n < 20\ \text{MeV}\), were calculated by using the MCNP\(^{5)}\) calculation code and the induced activities were calculated by the
DCHAIN-SP2001 calculation code with its related reaction cross section library.

COMPARISON BETWEEN EXPERIMENTAL AND CALCULATED RESULTS

Figure 6 shows the calculational to experimental (C/E) ratio of radioactivities for representative nuclides. Good agreement between calculated and experimental results within factor 2 was obtained for $^{24}\text{Na}$, $^{47}\text{Sc}$, $^{47}\text{Ca}$ and $^{54}\text{Mn}$ and within factor 5 was obtained for $^{42}\text{K}$, $^{43}\text{K}$, $^{51}\text{Cr}$ and $^{22}\text{Na}$. The productions of those nuclides were mainly due to the reaction by lower energy neutrons whose reaction cross sections were well estimated.

However, a large disagreement up to two orders of magnitude was observed for $^{28}\text{Mg}$, $^{54}\text{Sc}$, $^{52}\text{Mn}$, $^{7}\text{Be}$ and $^{56}\text{Co}$. Those nuclides were produced by spallation reactions, not produced by such reactions as (n,p), (n,np), (n,x) and (n,γ).

Large disagreements were also observed for other radioactive nuclides that were also produced by spallation reactions. The reason is that the compositions of chemical elements of their parent nuclides were not determined by chemical analysis. To estimate the induced activities for those radioactive nuclides by calculation, it is necessary to analyse the chemical composition of all the parent nuclides.

DISCUSSION

From the comparison between experimental and calculated results, it is clarified that the calculated results of the induced activities of radioactive nuclides, produced by spallation reactions, underestimate the experimental results. The reason is that these nuclides were very difficult to calculate by the NMTC/JAM calculation code system.

We have tried to estimate the induced activities of $^{7}\text{Be}$, as a nuclide produced by spallation reaction, by using the direct method. We have assumed that the parent nuclides of $^{7}\text{Be}$ were $^{12}\text{C}$, $^{40}\text{Ca}$, $^{16}\text{O}$ and $^{28}\text{Si}$. The induced activities of $^{7}\text{Be}$ were calculated by

![Figure 3. Experimental results of some radioactive nuclides of the limestone concrete.](image)
PRECISE ESTIMATION OF INDUCED ACTIVITIES IN CONCRETE SHIELDS

Ordinary (1 year)

![Figure 4](image1.png)

Figure 4. Experimental results of long-lived radioactive nuclides of the ordinary concrete.

![Figure 5](image2.png)

Figure 5. Block diagram of the calculation code system.
using the atomic density of parent nuclides, production cross sections, i.e. $^{12}$C(n,x)$^7$Be, $^{40}$Ca(n,x)$^7$Be, $^{16}$O(n,x)$^7$Be and $^{28}$Si(n,x)$^7$Be, which were mainly obtained from the JENDL-HE nuclear data library$^{(6)}$. The estimated cross sections of each reaction is listed in Table 1.

Figure 7 shows the calculated results and C/E values for all the sample positions. Very good agreement was obtained for all sample positions within factor 2, although large disagreement was obtained by the calculated results by using NMTC/JAM calculation code system.

### Table 1. Estimated cross sections of each reaction for $^7$Be production from the JENDL-HE nuclear data library.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Neutron (MeV)</th>
<th>Proton (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>100</td>
<td>1000</td>
</tr>
<tr>
<td>$^{12}$C</td>
<td>5</td>
<td>10</td>
</tr>
<tr>
<td>$^{14}$N</td>
<td>3</td>
<td>6</td>
</tr>
<tr>
<td>$^{16}$O</td>
<td>2</td>
<td>5</td>
</tr>
<tr>
<td>$^{27}$Al</td>
<td>0.2</td>
<td>6</td>
</tr>
<tr>
<td>$^{28}$Si</td>
<td>0.6</td>
<td>5</td>
</tr>
<tr>
<td>natSi</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{40}$Ca</td>
<td>0.5</td>
<td>7</td>
</tr>
<tr>
<td>$^{56}$Fe</td>
<td>0.16</td>
<td>5</td>
</tr>
</tbody>
</table>

Italics represent the estimated cross sections by using experimental data.

Factors 2–5 were obtained for the nuclides that were not mainly produced by the spallation reactions. However, for the nuclides that were produced by spallation reactions, the large disagreement up

### CONCLUSION

Induced activities of concrete samples were measured in the large concrete assembly irradiated by high-energy neutrons emitted from the tungsten target bombarded with 500 MeV protons. The calculated results, using the NMCT/JAM-MCNP-DCHAIN-SP2001 calculation code system, were compared to those of the experiment. Good agreements within
to two orders of magnitude was observed. Further study is necessary for the precise estimation of induced activities in concrete that were produced by the spallation reactions.

REFERENCES