

Depth Distribution of H-3, C-14 and Co-60 in Decommissioning of the Biological Shielding Concrete of KRR-2

S.B. Hong, H.R. Kim, K.H. Chung, K.H. Chung, J.H. Park
Korea Atomic Energy Research Institute
150 Deokjin-dong, Yuseong-gu, Daejeon, 305-353
S. Korea

Abstract

The depth distributions of the gamma emitting nuclides, H-3 and C-14 activities were characterized for the activated biological shielding concrete from a decommissioning of Korea Research Reactor-2 (TRIGA Mark-II) by using a commercially available tube furnace and a liquid scintillation counter. The HTO vapor and $^{14}\text{CO}_2$ gas generated from the tube furnace were trapped in HNO_3 and Carbosorb bubbler, respectively. The H-3 and C-14 activities were measured by the liquid scintillation counter. The specific activities of H-3 and C-14 were calculated by considering the concrete mass and the recovery factor of the tube furnace. The detection limits for H-3 and C-14 were 0.048 and 0.028 Bq/g respectively. The specific activity of the H-3 and C-14 tends to decrease exponentially as the depth of the concrete becomes deeper from the surface. In addition, the H-3 and C-14 activities were in good agreement with the Co-60 activities analyzed for the biological shielding concrete of KRR-2.

Introduction

Two decommissioning projects are being carried out at the KAERI (Korean Atomic Energy Research Institute), one for the Korea Research Reactors, KRR-1 and KRR-2, and another for the uranium conversion plant (UCP). In 1996, it was concluded that KRR-1 and the KRR-2 would be shut down and dismantled. The reactor decommissioning project was launched in January 1997 with the goal of a completion by 2008. The total budget of the project is 19.4 million US dollars, including the cost for the waste disposal and for the R&D. The decommissioning project work scope includes the dismantling of all the facilities and the removal of all the radioactive materials from the reactor site. After confirming the removal of all radioactivity, the site and buildings will be returned to the Korea Electric Power Company (KEPCO). At the end of 2005, dismantling of the KRR-2 was completed and all the radioactive waste from the KRR-2 was packed and stored in the KRR-2 reactor hall [1]. Decommissioning of a nuclear facility after its service life and its dismantling involves the necessity of a dismantling and handling of a considerable amount of radioactive waste. The most important remaining activated part is the bio-shielding concrete of the reactor. To reduce the volume of the waste and to optimize the dismantling of the bio-shielding concrete, the radioactivity and quantity of the activated part of the concrete should be evaluated as a part of decommissioning process.

The bio-shielding concrete was activated by a thermal and resonance neutron reaction during the operation of a reactor, thus a variety of radionuclides could have been generated in the activated

concrete, including H-3, C-14, Fe-55, Co-60, Ni-63, Cs-134, Cs-137, Eu-152 and Eu-154. However, most of the radioactivity comes from H-3 and C-14. In the bio-shielding concrete around a reactor core, C-14 mainly comes from neutron activation reaction from $^{14}\text{N}(\text{n}, \text{p})^{14}\text{C}$, and $^{17}\text{O}(\text{n}, \alpha)^{14}\text{C}$ may have a slightly higher contribution due to a higher concentration of oxygen in the concrete. The main contribution of H-3 in the concrete comes from the neutron activation reaction $^6\text{Li}(\text{n}, \alpha)^3\text{H}$, with a very small amount of H-3 probably coming from the reaction $^2\text{H}(\text{n}, \gamma)^3\text{H}$ and $^3\text{He}(\text{n}, \text{p})^3\text{H}$. ^6Li has a high neutron activation cross section(940 barns). In concrete, C-14 exists as carbonate or carbon, and H-3 as HT or HTO in the pore and gaps of the concrete. H-3 and C-14 are pure beta emitters, and the energies of their beta particles are relatively low($E_{\beta\text{max}}$ H-3=18.6keV, $E_{\beta\text{max}}$ C-14=156.5keV), thus the concrete needs to be decomposed to separated H-3 and C-14 from the matrix. The radioactivity of the concrete was measured by using a liquid scintillation counter(LSC), due to its low energy[2-5].

This research presents a evaluation of the radionuclide specific inventory in the bio-shielding concrete of KRR-2, and analysis of the correlation between the gamma emitters and beta emitters. The radioactivity of gamma emitters is measured by using HPGe detector with MCA(Multi Channel analyzer), and H-3 and C-14 are measured by an oxidation combustion method in a tube furnace. The correlations between gamma emitters and beta emitters such as H-3 and C-14 are analyzed for the activated concrete from the KRR-2.

Experimental

Sample Preparation

The activated part of the bio-shielding concrete of the KRR-2 is mixed with magnetic aggregates, and the density is $2.8 \sim 3.4 \text{ g/cm}^3$. For a characterization of the bio-shielding concrete, three works were carried out. The first work was an understanding of the exact shape and size of the bio-shielding concrete and the second was a measurement of the physical properties such as density. Finally a matrix sampling on the surface and samplings along the depth of the concrete were carried out. From the measured radioactivity of the samples, a mapping of the surface radioactivity was carried out and it was extended to 3 dimensional diagrams with a general dependency of the radioactivity along the depth. More than 67 samples of the concrete were collected from around the bio-shield concrete. The most highly activated part of the sample was located around the thermal column because of the proximity to the reactor core and the thermal column is filled with a graphite which was used for a neutron moderator or reflector in the reactor. This paper presents the measurement and correlation results for the gamma emitters and beta emitters in the activated concrete around the thermal column. Core drilling machines were used to take samples at a distance of 10 cm apart around the thermal column in the KRR-2.

Measurement & Analysis Procedures

The gamma-ray emitting nuclides were measured by a HPGe detector(Canberra) connected to a multi-channel analyzer, the full width at half-maximum resolution of 1.8 keV measured at 1.33MeV(^{60}Co) and a relative efficiency of 47.1%. The sample is directly determined by correcting a self-absorption effect. The concrete samples were crushed into powder and poured

into cylindrical vials for a measurement. The weight of the analyzed sample was around 150g and its measuring time was 3600s.

The radioactivity of H-3 and C-14 are determined by combusting the concrete sample in a purpose-designed tube furnace using a predetermined heating cycle. The bubbler solution was mixed with a scintillator(Goldstar) and each sample was counted using a liquid scintillating counter. The tube furnace(Pyrolyser 6TM) was purchased from RADDEC^{LTD}(National Oceanography Centre, UK). A QuantulusTM 1220 liquid scintillation counter was used for a measurement of the H-3 and C-14 activities. The tube furnace was comprised of six silica tubes that pass through the furnace. Two furnace zones are separated by a 15 cm insulated gap providing thermal isolation of the sample-zone from the catalyst-zone. For the concrete sample, 5 g samples were placed on the aluminum boat that was transferred into the middle of the sample-zone in the silica-glass work tube. The combustion products in the sample-zone of the furnace were passed through a 0.5% Pt-alumina catalyst (10 g) in the catalyst-zone, which was heated to 800 . Compressed pure air was passed over the sample, which was progressively heated to 500 by using a predetermined ramping cycle. When the temperature of the sample-zone reached to 500 , additional oxygen was supplied to ensure complete oxidization of the sample. The flow rate of compressed air and oxygen was approximately 0.25 ~ 0.3 L/min. A schematic diagram of the oxidization method is given in Figure 1. After the oxidization process, the tritiated water was trapped in HNO₃(0.1 M 20 ml) bubblers, and ¹⁴CO₂ was trapped in Carbosorb(20 ml) bubbler respectively. To measure the radioactivity of H-3 and C-14, 8 ml of the solution was transferred to a 20 ml LSC vial, and 12 ml of Goldstar scintillation cocktail was added and mixed for measuring the H-3 concentration. Five ml of the solution and 12 ml of the Goldstar cocktail were added and mixed in the 20 ml LSC vial for measuring the C-14 concentration.

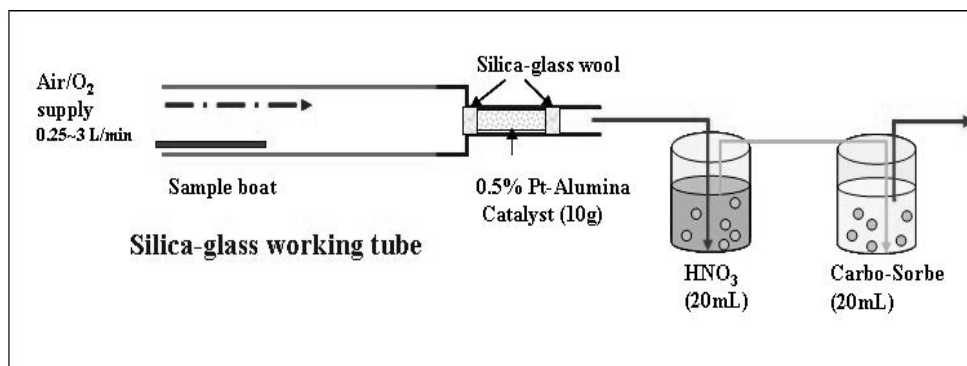


Figure 1. Schematic diagram of the oxidization method by using the tube furnace.

The activities of H-3 and C-14 were measured by a liquid scintillation counter. Low-energy and high-energy windows were used for the measurement of H-3 and C-14, respectively. The quenching level was measured by the external standard method SQP(E), and the counting efficiency was calculated by using standards at different quenching levels was given in Figure 2. To determine the recovery of the furnace, the standard source(165 Bq/g; H-3, 185Bq/g; C-14) was sparked on the filter paper(Whatman 42, ash less 0.01%). The average furnace recovery of H-3 and C-14 obtained from a replicated analysis was around 90% and 95% respectively.

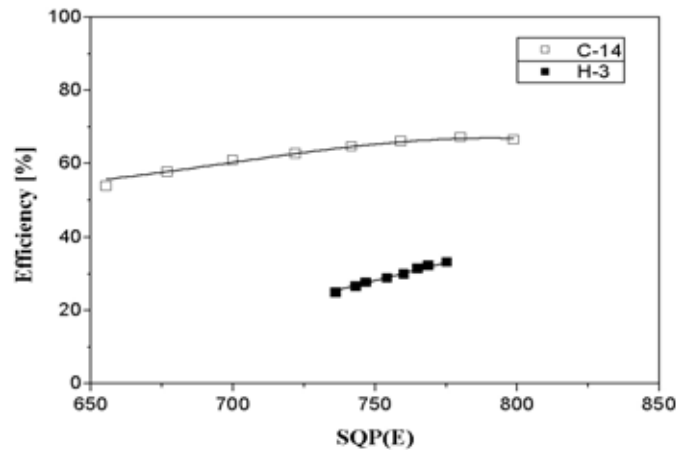


Figure 2. Counting efficiency of H-3 and C-14 for different quenching levels

Results

The analytical results of the activities of the gamma emitting nuclides were measured and the peak concentration was 150 Bq/g. Co-60, Cs-134, Eu-152 and Eu-154 were identified as the radionuclides in the activated concrete of KRR-2. The most gamma activity came from Co-60 however the Cs-134, Eu-152 and Eu-154 nuclides had a very low activity around the highly activated part of the concrete. The specific activity of the gamma emitting nuclides tended to decrease exponentially as the depth of the concrete increased.

The activities of the H-3 and C-14 in the bubbler and sample were calculated by using the formula below. The results can be achieved by using the registry file of the LSC and the data was calculated by using the LSC+ software which was developed by RADDEC^{LTD}.

$$A_B = \frac{C - B}{60} \times \frac{100}{E} \times \frac{1}{m} \quad \text{eq.1)}$$

where A_B = the activity concentration in the bubbler (Bq/g)

C = the sample count rate (CPM)

B = the background count rate (CPM)

E = the counter efficiency (%)

M = the mass of bubbler solution taken for analysis (g)

$$A_S = A_B \times \frac{(m_f - m_t)}{m_s} \times \frac{100}{R} \quad \text{eq. 2)}$$

where A_S = the activity concentration in the sample (Bq/g)

m_f = the final bubbler mass (g)

m_t = the bubbler tare mass (g)

m_s = mass of sample taken (g)

R = furnace recovery (%)

The results for H-3 and C-14 in the concrete are given in Figure 3 depending on the depth. Each sample was measured for 30 minutes and three times by using LSC, the average of the three measurements being calculated by using the LSC+ software. The H-3 and C-14 levels were measured as 1680 Bq/g and 0.25 Bq/g in the most highly activated part of the concrete, respectively. Based on the measurement results, the correlation between H-3, C-14 and Co-60 was evaluated and it showed that the depth distribution of the activities was decreased with the same trends in the biological shielding concrete of KRR-2. The correlation coefficients for the calculated equation were 0.983 and 0.885. Figure 4 shows the correlation between H-3, C-14 and Co-60.

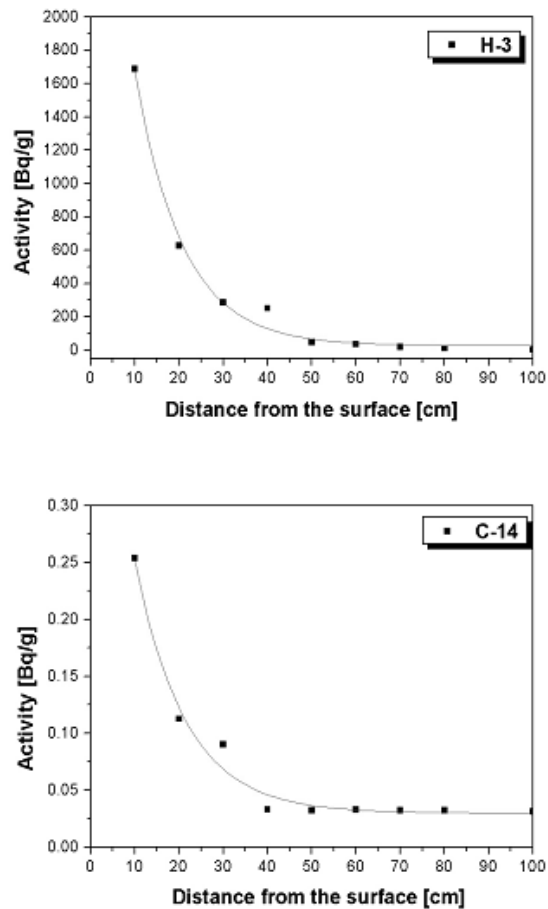


Figure 3. Analytical results of the H-3 and C-14 in the activated concrete of the KRR-2.

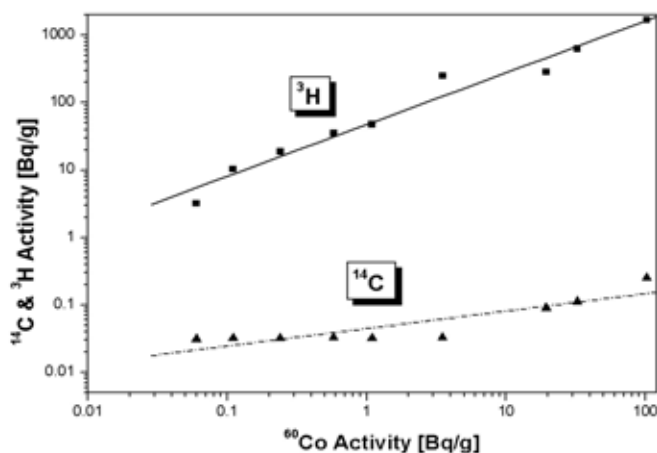


Figure 4. The correlation between gamma emitter (Co-60) and beta emitter (H-3 and C-14) in activated concrete from the KRR-2.

Conclusion

The bio-shielding concrete was activated by a thermal and resonance neutron reaction. The pure beta emitting nuclides such as H-3 and C-14 were measured. In order to identify the correlations between H-3, C-14 and Co-60 in the activated concrete of KRR-2, and the analytical measurement procedures were set-up and applied. The correlations H-3 and C-14 activities were in a good relation with the Co-60 activities for the bio-shielding concrete. The measured results and procedures could provide important information for the waste management of a reactor decommissioning project. These results provide a solution for estimating the inventory of the activated solid waste of KRR-2.

References

1. K. W. Lee, S. K. Park, S. B. Hong, U. S. Chung and J. H. Park, "Experiences of the dismantling on non-activated and activated bio-shielding concrete of KRR-2," Proceedings of the 7th joint workshop between China and Korea," pp. 271-281(2006).
2. Akira Endo, Yasunori Harada, Katsuya Kawasaki and Masamitsu Kikuchi, "Measurement of depth distributions of H-3 and C-14 induced in concrete shielding of an electron accelerator facility," Applied Radiation and Isotopes, 60, pp. 955-958(2004).
3. Xiaolin Hou, "Rapid analysis of ^{14}C and ^3H in graphite and concrete for decommissioning of nuclear reactor," Applied Radiation and Isotopes, 62, pp. 871-882(2005).
4. J. P. Krasznai, "The radiochemical characterization of regular- and high-density concrete from a decommissioning reactor," Waste Management, Vol. 13, pp. 131-140(1993).
5. H. J. Ahn, H. N. Lee, S. H. Han, "Simultaneous assay of ^{14}C and ^3H in Evaporator bottom by Chemical oxidation method," J. of the Korean Radioactive Waste Society, vol.3(3), pp193-200(2005).