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The radioactivity analysis of ^{14}C and ^{3}H in graphite from the dismantled korea research reactor and its dose estimation

H.R. Kim, S.B. Hong, W. Lee, G.H. Chung, G.S. Choi and C.W. Lee

Korea Atomic Energy Research Institute, 305-600 Daejeon, Korea

Abstract. A high temperature combustion method was used to analyze the ^{14}C and ^{3}H activities in graphite and the dose assessment was carried out to determine the clearance in the conservative way. By this method, the ^{3}H and ^{14}C were simultaneously trapped in the nitric acid and carbosorb, respectively. Accordingly, the sample preparation time for the measurement was reduced to the half. The combustion temperature was more than 800 degrees in centigrade for obtaining total tritium and ^{14}C in the sample. The combustion ratio was about 99% on the graphite sample with the weight of 0.1 g. Minimum detectable activity was 0.05 Bq/g for the ^{14}C and 0.15 Bq/g for the ^{3}H at the same background counting time. The recoveries from the combustion furnace were around 100% and 90% in ^{14}C and ^{3}H, respectively. The radioactivity were 2,530 ∼3,160 Bq/g in ^{14}C and 1,700 ∼2,040 Bq/g in ^{3}H at this experiment. The experimental uncertainty was less than 6% in both radionuclides where the furnace recovery was dominant factor. An individual effective dose from beta and gamma radionuclides was estimated by consideration of the scenario of inhalation, ingestion and external exposure. ^{60}Co, the radioactivity of which was measured by using HPGe detector, had a predominant effect in estimating the effective dose. The estimation showed that the graphite wastes from the dismantled research reactor should be disposed of as a low level radioactive waste rather than clearance.

1. INTRODUCTION

A nuclear installation, like research reactors, is shut down and decommissioned when its life is over and its operation is stopped. Actually, the Korea Research Reactor (KRR-2) was completely dismantled after an operation of more than 30 years. Therefore, various radioactive wastes such as concrete, metal, graphite and others were generated during its dismantling. Of these dismantling wastes, especially, the irradiated graphite has become a common part of the decommissioning activities throughout the world for its handling, conditioning and disposal. The graphite in the thermal column had been irradiated by neutrons for more than 30 years and graphite radioactive waste with a weight of 3,126 kg was generated during the dismantling. It is predicted to have various radionuclides such as ^{3}H, ^{14}C, ^{36}Cl (beta nuclides), and ^{60}Co, $^{134}Cs4$, ^{152}Eu (gamma nuclides). Of these radionuclides, the tritium and radioactive carbon have been a matter of concern from the aspect of an internal exposure like an inhalation or ingestion. The ^{14}C and ^{3}H radionuclides are known to have a high mobility in the environment. Carbon remains for a few days or weeks and the tritium remains for about 10 days in the human body. In the present study, the graphite from the dismantled thermal column was sampled from the graphite block and its ^{14}C and ^{3}H radioactivity was analyzed by a liquid scintillation counting (LSC) method. Also, a dose assessment for ^{14}C and ^{3}H is discussed from the point of view of a clearance for the determination of a reuse and recycling.

2. MATERIALS AND METHODS

2.1 Sampling and radioactivity analysis

The graphite block used as a moderator in the KRR-2 forms the vertical and horizontal thermal columns as presented in Fig. 1. It is composed of carbon, hydrogen, oxygen, nitrogen and other components. Especially, the portion of the carbon is more than 99.9% while the hydrogen and others are less than 0.001%. The graphite block has a geometrical size with a width of 3,350 mm, a length of 1,220 mm

and a height of 1,200 mm. The sampling zone is divided into three parts radially and azimuthally from the center of the reactor core. The graphite sample with a weight of 0.1 g was taken for a complete oxidization in a high temperature of 800 °C. A pre-treatment by using a chemical procedure and a combustion of the samples was carried out to analyze the radioactivity of ^{14}C and ^{3}H of the dismantled graphite waste [1, 2]. Nitric acid with a concentration of 0.1 M and carbosorb were pipetted at 20 mL each and they were poured into the corresponding bubblers. Then, the bubblers were connected to the quartz tubes of the combustion furnace with a 0.5% Pt-alumina catalyst by using rubber tubes. The graphite samples with a weight of 0.1 g were burned at a maximum temperature of 800 °C for 440 minutes. During the burning process, the ^{14}C and ^{3}H of the graphite samples were trapped in the nitric acid and carbosorb of the bubbler, respectively. The carbosorb including the ^{14}C was mixed with a scintillation solution at a ratio of 5 to 15 in a vial while the nitric acid including the ^{3}H was mixed with a scintillation solution at a ratio of 8 to 12. These vials were placed into the LSC (Quantlus 1220, Walac) and finally their radioactivity was measured. Fig. 2 shows the general experimental system for measuring the radioactivity.

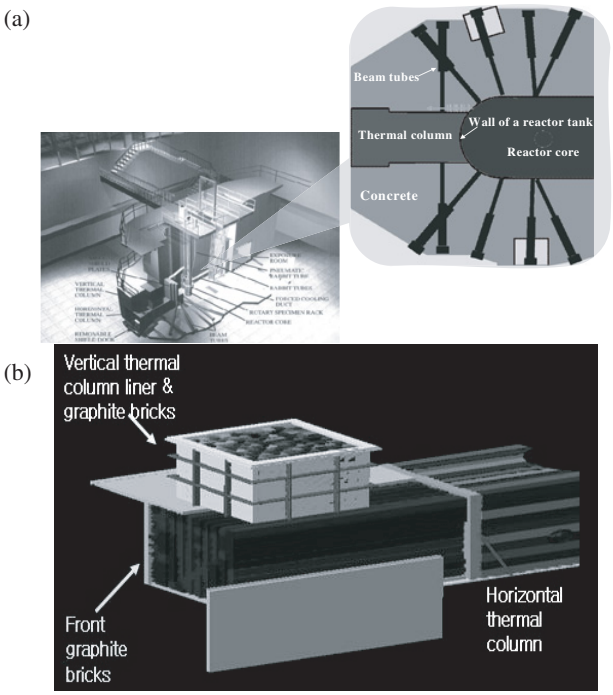

Figure 1. The Korea Research Reactor-2 (KRR-2) and the thermal column of the graphite block. (a) KRR-2, (b) The thermal column of the graphite block.

The specific activities of the ^{14}C and ^{3}H were $2,530\,Bq/g \sim 3,160\,Bq/g$ and $1,700\,Bq/g \sim 2,040\,Bq/g$, respectively, for the samples where the combustion ratio was $99.0\% \pm 0.0202\%$. These numerical figures are much larger than the limit of a concentration available for a self-disposal where they are 1 Bq/g for the ^{14}C and 100 Bq/g for the ^{3}H, respectively [3]. Also, the minimum detectable activities (MDA) were calculated to be 0.05 Bq/g for the ^{14}C and 0.15 Bq/g for the ^{3}H by using Curie's equation for the samples, respectively [4]. On the other hand, because the measurement errors can be assumed to be independent of each other and the presentation of the radioactivity has a product form as presented in equation (1), its combined relative uncertainty can be expressed in the form of equation (2)

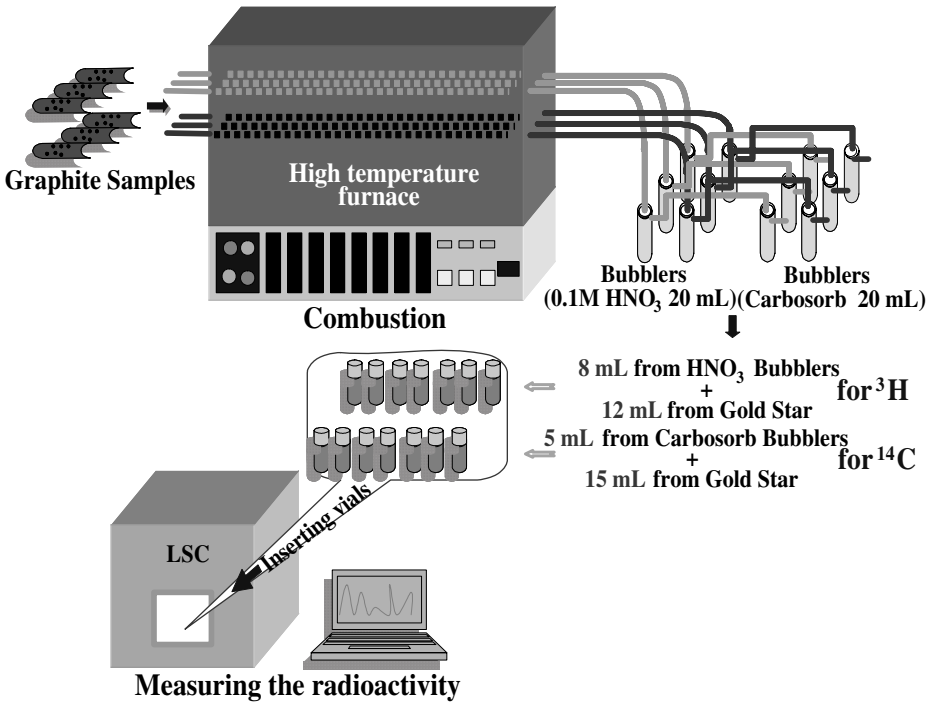

Figure 2. The experimental system with a commercialized furnace and a LSC for measuring the radioactivity.

through some mathematical process [1, 5–7].

$$A_S = \frac{C-B}{60} \times \frac{100}{E} \times \frac{1}{m} \times \frac{m_f - m_t}{m_s} \times \frac{100}{R} \tag{1}$$

$$\left(\frac{U_{A_S}}{A_S}\right)^2 = \left(\frac{U_{C-B}}{C-B}\right)^2 + \left(\frac{U_E}{E}\right)^2 + \left(\frac{U_m}{m}\right)^2 + \left(\frac{U_{m_f-m_t}}{m_f-m_t}\right)^2 + \left(\frac{U_{m_s}}{m_s}\right)^2 + \left(\frac{U_R}{R}\right)^2 \tag{2}$$

The relative uncertainty of a sample weighing, mass of a bubbler solution taken for an analysis and the difference between the final bubbler mass and the bubbler tare mass were negligible, less than 0.005%, in this experiment. The relative uncertainty of the decay correction and certified ^{14}C standard solution (as quoted on certificate) were 0.2% and 0.67% respectively. The relative uncertainty of the furnace recovery was 4.9% in maximum where the furnace recovery, which was a dominant factor for the uncertainty, 100% ± 2.5% and 90% ± 4.9% for the ^{14}C and ^{3}H each and that of the liquid scintillation counter calibration curve was 1.0%. Therefore, the combined relative uncertainty was calculated to be about 5.1% in maximum, which corresponded to the case of the ^{3}H. In fact, this recovery considerably depends on the condition of the Pt catalyst, temperature and burning duration. Hence, it is thought that this uncertainty can be remarkably reduced by carrying out an exchange of the catalyst, and the maintenance of the suitable temperature and duration.

2.2 Dose estimation

An individual effective dose estimation is practically important to determine whether the graphite waste from the dismantling is self disposed of or disposed in a low/intermediate level disposal site. Because ^{14}C and ^{3}H are pure beta emitters, an internal exposure like an inhalation or ingestion has an

influence on the dose estimation. The dose estimation is carried out by using the IAEA's application of exemption principles to the recycle and reuse of materials from nuclear facilities. The dose estimation equations [8–10] are represented by the Eqs. (3) and (4) where the dose conversion factors for an inhalation were $6.4 * 10^{-12}$ Sv/Bq and $1.7 * 10^{-11}$ Sv/Bq for the ^{14}C and ^{3}H, respectively, and the dose conversion factors for an ingestion were $5.6 * 10^{-10}$ Sv/Bq and $1.7 * 10^{-11}$ Sv/Bq for the ^{14}C and ^{3}H, respectively.

$$H_{INH} = V\,t\,DF_{INH}\,W(C_d\,C_w + C_s RFTF_{INH}) \tag{3}$$

$$H_{ING} = t\,DF_{ING}\,W(IC_{ING} + I2\,TF_{ING}C_s) \tag{4}$$

First, calculating the dose from ^{14}C gave 0.17 uSv/y $\sim$ 0.21uSv/y for an inhalation and 124 uSv/y $\sim$ 155 uSv/y for an ingestion, respectively. The dose from ^{3}H was computed to be 0.30 uSv/y $\sim$ 0.37 uSv/y for an inhalation and 2.53 uSv/y $\sim$ 3.04 uSv/y for an ingestion, respectively. So, the internal dose by the two radionuclides was between 127 uSv/y and 159 uSv/y. These figures are much higher than the legal limit of an annual dose rate for a self-disposal or clearance, 10 uSv/y [3]. Furthermore, if the external exposure effect by gamma radionuclides like ^{60}Co, ^{134}Cs and ^{152}Eu included in the graphite samples is added, the total dose will be higher.

3. RESULTS AND CONCLUSIONS

The radioactivity of ^{14}C and ^{3}H in the graphite samples from the KRR-2 were analyzed by using a high temperature combustion furnace and the LSC method. The radioactivity of the ^{14}C was a bit higher than that of the ^{3}H where the activity ratio of ^{14}C to ^{3}H was about 1.5. The dose estimation indicated that the internal exposure by an ingestion was a few hundred times higher than that by an inhalation in the case of ^{14}C while that by an ingestion was a few times higher than that by an inhalation in the case of ^{3}H. In this estimation, the ingestion from ^{14}C contributes about 98% to the total internal dose. Also, the dose from the present analysis was much higher than the legal limit of an annual dose for a self-disposal, 10 uSv/y, although the dose from the other radionuclides like gamma radionuclides was not included. Consequently, the dose estimation due to the ^{14}C and ^{3}H radionuclides revealed that the graphite waste from the thermal column of KRR-2 had a difficulty for a clearance from the analysis results. It was concluded that the graphite blocks should actually be disposed of as a low or intermediate level radioactive waste after a further dose estimation including the effects from the gamma radionuclides for the public or the workers.

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NOMENCLATURE

As = The activity concentration in the sample (Bq/g)
B = The background count rate (CPM)
C = The sample count rate (CPM)
C_d = The concentration of respirable dust in air (0.001 g/m^3)
C_s = The concentration present as surface contamination (1 Bq/cm^2 in this analysis)
C_w = The concentration of the radionuclide (Bq/g)
C_{ING} = The concentration of the radionuclide (Bq/g)
DF_{ING} = The dose conversion factor from ingestion (Sv/Bq)
DF_{INH} = The dose conversion factor from inhalation (Sv/Bq)

E = The counter efficiency (%)
H_{ING} = The committed effective dose equivalent from one year's intake by ingestion (Sv)
H_{INH} = The committed effective dose equivalent from one year's intake by inhalation (Sv)
I = The rate of secondary ingestion of removable contamination (0.01 g/h in this analysis)
I2 = The rate of secondary ingestion of removable surface contamination (10^{-4} m^2/h in this analysis)
m = The mass of bubbler solution taken for analysis (g)
mf = The final bubbler mass (g)
ms = Mass of sample taken (g)
mt = The bubbler tare mass (g)
R = Furnace recovery (%)
RF = The resuspension factor for surface activity (10^{-6} m^{-1} in this analysis)
t = The duration of exposure for the individual (h)
TF_{ING} = The transfer factor for the ingestion of surface activity (0.01 in this analysis)
TF_{INH} = The transfer factor for the inhalation of surface activity (10^{-6} in this analysis)
U_{As} = The uncertainty of the activity concentration in the sample (Bq/g)
$U_{C\text{-}B}$ = The uncertainty of the net count rate (CPM)
U_E = The uncertainty of the counter efficiency (%)
U_m = The uncertainty of the mass of bubbler solution taken for analysis (g)
$U_{mf\text{-}mt}$ = The uncertainty of the difference of the final bubbler mass and the bubbler tare mass (g)
U_{ms} = The uncertainty of mass of sample taken (g)
U_R = The uncertainty of furnace recovery (%)
V = The ventilation rate (1.2 m^3/h in this analysis)
W = The fraction of the material handled (1.0 in this analysis)

References

[1] I. Croudace and Jung.-Suk. Oh, Analysis of total tritium and ^{14}C in solid samples, GAU/RC/2022 (2005).
[2] Hee Reyoung Kim, et al., "The Radioactivity Analysis of ^{14}C in the Graphite Samples from the Dismantled KRR-1&2 Sites by a high temperature furnace and a LSC", Proceedings of the 11^{th} International Conference on Environmental Remediation and Radioactive Waste Management ICEM07, September 2-6, 2007.
[3] MEST Notice 2001-30 (2001).
[4] IAEA, Measurement of Radionuclides in Food and the Environment, Technical Report Series No. 295, Vienna, 1989.
[5] Robert J. Moffat, "Describing the Uncertainties in Experimental Results", Experimental Thermal and Fluid Science, Vol. 1, pp. 3-17, 1988.
[6] James D. Jenkins, Measurement Uncertainty Analysis Fundamentals, Quametec Corporation, 2004.
[7] Hugh W. Coleman and W. Glenn Steele, Experimentation and Uncertainty Analysis for Engineers, John Willey and Sons, 1989.
[8] UNSCEAR, Sources and Effects of Ionizing Radiation, Annex A: Dose Assessment Methodologies, UNSCAER 2000, United Nation Scientific Committee on the Effect of Atomic Radiation, (2000).
[9] IAEA Safety Series No. 111-P-1.1, Application of Exemption Principles to the Recycle and Reuse of Materials from Nuclear Facilities, IAEA, 1992.
[10] IAEA Safety Series No. 115, International Basic Safety standards for Protection Against Ionizing Radiation and for the Safety of radiation Sources, IAEA, 1996.