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Development of the Dual Counting Method and Internal Dose Assessment Method for Tritium and Carbon-14 at Nuclear Power Plants

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1. Introduction

At pressurized heavy water reactors (PHWRs), carbon-14 is generally produced by the irradiation of oxygen present as impurities in the moderator system from $^{17}O(n,\alpha)^{14}C$ reaction. Because carbon-14 release from PHWRs takes place mostly as carbon dioxide ($^{14}CO_2$), in the case of carbon-14 release to the work area, it is easily inhaled into the body of radiation workers. Most inhaled carbon-14 is rapidly exhaled from the worker's body, but a small amount of carbon-14 remains inside the body and is excreted by urine. Internal dose assessment for carbon-14 is conducted using the measurement results of cabon-14 in urine samples of radiation workers since cabon-14 is a low energy beta emitter.

In this study, the origin of cabon-14 and its metabolism through inhalation were reviewed for internal dose assessment of carbon-14 at PHWRs. In particular, the method of dual analysis of tritium and carbon-14 in urine samples was developed with the use of Liquid Scintillation Counters (LSCs) and methods for tritium measurements at the Wolsong nuclear power plants (NPPs). In addition, methods for determination of intake and internal dose assessment were established based on the measurement results of carbon-14 and its excretion rate data.

2. Methods and Results

2.1 Dual analysis of tritium and carbon-14

In development of the method of dual analysis of tritium and carbon-14, measurement and analysis were performed with LSC and both artificial and real urine samples of radiation workers [1,2]. Prior to measurements, sensitivity analysis was conducted for each parameter, which can affect the counting results. The important parameters include the background of counting vials, stabilization interval of samples, change of efficiency depending on the mixture proportion of sample and cocktail, irradiation interval of external sources, and Quenching Index Parameter depending on samples. After sensitivity analysis, effective channels of LSC were determined through the optimization process, which increases the effective count's maximum and reduces the background minimum. As a result, the effective channels were set from Channels 1 to 9 for single and dual analysis of tritium, from Channels 4 to 85 for single analysis of carbon-14, and both from Channels 30 to 85 and Channels 40 to 85 for dual analysis of carbon-14 [1].

2.2 Application of dual analysis to NPPs

A reliability test for the developed method of dual analysis was conducted using standard samples made for the use of verification. To make standard samples, tritium was mixed at a level of radioactivity 100 times higher than that of carbon-14, with consideration of the practical level of radioactivity in urine samples at NPPs. As a result of the reliability test, the results demonstrated good performance, showing that the mean values of analysis for tritium and carbon-14 were almost similar to the calculated values, within 10 % for tritium and 5 % for carbon-14. Table 2 displays the verification results of the analyses for tritium and carbon. The standard deviation of the measured values was regarded as appropriate, and was shown to be within 8 ~ 10 % of average measured values [1,3].

As a result of the application of dual analysis to urine samples of radiation workers at Wolsong NPPs, it was found that measurement results of tritium activity almost corresponded to previous measurement results of tritium activity. Thus, the validity of the dual analysis method was indirectly demonstrated. In the meantime, there was the phenomenon of crossover, the interchange of sections between tritium and carbon-14 counts, for the results of carbon-14 analysis when Channels 4 ~ 85 were set for carbon-14 analysis; this caused overestimation of carbron-14 counts. On the other hand, the phenomenon of crossover of tritium counts was properly eliminated when Channels 30 ~ 85 and Channels 40 ~ 85 were set for carbon-14 analysis. In addition, it was found through dual analysis that counting results of carbon-14 for urine samples of radiation workers at PHWRs were always lower than MDA, and that the likelihood of internal exposure to carbon-14 is extremely low [1,4]. Several measurements of tritium and carbon-14 activity for actual urine samples of radiation workers at PHWRs were conducted applying the developed method for dual analysis of tritium and carbon-14 [1].

The dual counting method for tritium and carbon-14 in workers' urine was carried out for radiation workers who participated in tasks for which high radiation exposure was expected, such as the open process of steam generators during the planned maintenance period at PWRs. As a result, it was found that tritium activity in urine samples of radiation workers at PWRs indicated almost MDA levels, much lower than those at Wolsong NPPs (PHWRs). Thus, this result demonstrated indirectly that the likelihood of internal exposure to tritium at PWRs is extremely low. Furthermore, there was no detection of carbon-14. This result confirms that there was no carbon-14 in urine samples of radiation workers at PWRs and that internal exposure to carbon-14 is unlikely to occur [1,4].

2.3 Carbon-14 metabolism and its excretion rate

The International Commission on Radiological Protection (ICRP) provided some information about carbon-14 in ICRP publication 10, stating that inhaled CO_2 remains inside the body for a 0.4-day retention period, and that 30 % of inhaled CO_2 is deposited in the bones. In ICRP publication 30, results were presented that showed that 99 % of inhaled CO_2 exhibit two short-term behaviors that have a 5-minute or a 60-minute retention period and that the final 1 % of residual carbon-14 remains inside body for 60,000 minutes (approximately 40 days). This carbon metabolism is based on test results on animals. In the meantime, ICRP publication 54 (1983) and publication 78 (1997) do not provide detailed information about carbon-14 metabolism or give any guidance on internal exposure monitoring. According to ICRP publications, the central compartment of radioactive carbon dioxide ($^{14}CO_2$) inhaled by breathing or ingested by diet is the behavior of CO_2/HCO_3^- [5].

In Canada, measurement experiments have been performed to examine the excretion rate of carbon-14 for volunteers who have inhaled low levels of the substance, and for whom the density of inhaled $^{14}CO_2$ was known; these experiments were done to estimate the internal exposure of NPP workers to inhaled CO_2. It was found that radiocarbon excreted was present as bicarbonate in urine and that it had retention periods of 0.4, 1.2, and 40 days, respectively. Based on this metabolism data, the Canadian Nuclear Safety Commission, the nuclear regulatory body, organized the Working Group on Internal Dosimetry and provided bioassay guideline for carbon-14 inhalation [6,7].

2.4 Internal Dose Assessment

The dose rate (rem/d) to a tissue of mass m (g) resulting from radioactivity Q (μ Ci) of carbon-14 is given by Eq. 1 [1,7,8]. Here, E (MeV) is the mean energy of the beta emission. The committed effective dose (rem) resulting from an initial radioactivity Q_o (μ Ci) is described in Eq. 2. Inhaled carbon-14 is distributed throughout all soft tissues of the body. Thus, the committed effective dose, resulting from the mean energy 0.049 MeV and the soft tissue mass of Reference Man 63,000 g, is given by Eq. 3. For a urine sample of concentration C (μ Ci/L) obtained at time t after single or acute intake, the intake can be estimated by Eq. 4. Here, "1.4 L" is the nominal daily excretion of urine. The final committed effective dose is then given by Eq. 5, which can be described in Eq. 6 using SI unit (mSv).

$$Dose\_Rate = \frac{51.2 \times E \times Q}{m} \quad \text{(Eq. 1)}$$

$$CED = \frac{51.2 \times E \times Q_o}{m} \int R(t) e^{\frac{-0.693t}{T_{1/2}}} dt \quad \text{(Eq. 2)}$$

$$CED = 5.0 \times 10^{-5} \times Q_o \ \text{[rem]} \quad \text{(Eq. 3)}$$

$$Intake = \frac{1.4 \times C}{E_u(t)} \quad \text{(Eq. 4)}$$

$$CED = \frac{7.0 \times 10^{-5} \times C}{E_u(t)} \ \text{[rem]} \quad \text{(Eq. 5)}$$

$$CED = \frac{25.9 \times C}{E_u(t)} \ \text{[mSv]} \quad \text{(Eq. 6)}$$

3. Conclusions

In this study, carbon-14 metabolism was investigated to establish a methodology for internal dose assessment of carbon-14 inhalation. Furthermore, a method for dual analysis of tritium and carbon-14 in urine samples of NPP workers was developed. Finally, the procedures for measurement and dose calculation of tritium and carbon-14 activity were provided based on the experiment and investigation results.

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