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Preliminary Study on the Internal Dosimetry Program for Carbon-14 at Korean CANDU Reactors

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1. Introduction

More strict radioactive regulations are applied to Korean nuclear power plants (NPPs) since ICRP-60 recommendation for radiation protection published by the International Commission on Radiological Protection (ICRP) in 1990 is legislated and has been enforced since 2003. Sixteen PWR reactors and four Canada deuterium-uranium (CANDU) reactors are currently in operation in Korea. In particular, carbon-14 and tritium concentrations are significantly higher at CANDU reactors compared to PWR reactors and this increases the risk of internal radiation exposure to workers at CANDU NPPs. Thus, it is necessary to estimate the exact amount of internal radiation exposure to workers for radiological protection at CANDU reactors. However, Korean CANDU reactors do not have a standardized set of regulations and internal dosimetry method for carbon-14. In this paper, the current dosimetry method for carbon-14 is analyzed for the establishment of internal dosimetry for carbon-14 at domestic NPPs. In addition, the production mechanism and characteristics of carbon-14 is described to understand its physical and chemical specification and environmental behavior.

2. Production and Characteristics of Carbon-14

2.1 Production of Carbon-14

Carbon-14 is a low-energy pure beta emitter with a half life of 5730 years. According to Korean Final Safety Analysis Report (FSAR), the annual amount of carbon-14 produced in a NPP is estimated 8.2 Ci/y for Yonggwang PWRs No. 3&4, 5.8 Ci/y for Ulchin PWRs No. 3&4 and 370 Ci/y for Wolsong CANDU reactors. These results showed that production yields of carbon-14 in CANDU reactors are 40~50 times higher than those of PWRs. Figure 1 illustrates the annual amount of carbon-14 produced in a CANDU NPP [1~3].

During operation, CANDU reactors produce carbon-14 primarily through neutron reaction with oxygen, nitrogen and carbon. These elements are present in the fuel, annulus gas and moderator. Table 1 shows the principal nuclear reactions forming carbon-14 in a CANDU reactor [4].

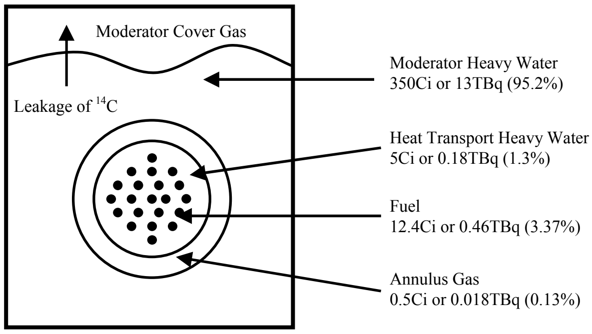

Figure 1. Production of Carbon-14 in a Typical CANDU 6 Reactor

Table 1. Principal Reactions Forming Carbon-14

Target nuclide	Natural abundance	Reaction	Thermal neutron cross section
^{17}O	0.037%	$^{17}O(n,\alpha)^{14}C$	0.235 barn
^{14}N	99.63%	$^{14}N(n,p)^{14}C$	1.820 barn
^{13}C	1.11%	$^{13}C(n,\gamma)^{14}C$	0.009 barn

2.2 Characteristics of Carbon-14

Carbon-14 is a long-lived radionulclide ($T_{1/2}$: 5730 years) which decays according to the following equation by the emission of a moderately-weak beta minus particle to stable nitrogen [5]. The maximum energy of the carbon-14 is 156 keV with a beta range in tissue of 0.28 mm. Thus, carbon-14 gives no external hazard to the skin; however, intakes or inhalation of carbon-14 create a potential occupational hazard.

$$^{14}_{6}C \rightarrow \beta^{-} + ^{14}_{7}N + \bar{\nu}$$

Carbon-14 in the moderator and heat transport systems is present mainly as soluble CO_2/HCO_3^-, and continually removed from these systems together with other contaminants, by cycling through resin filters. However, sufficient quantities of airborne $^{14}CO_2$ release into the workplace during maintenance on these systems and create a significant hazard to maintenance workers.

3. Internal Dosimetry for Carbon-14

3.1 Dosimetry for $^{14}CO_2$

All radiation workers take a regular urine bioassay program at Canada Ontario Power Generation (OPG) NPPs to estimate the internal radiation exposure. Energy discrimination of the liquid scintillation counter (LSC) response makes both carbon-14 and tritium to be determined separately in the same urine samples since the energies of most carbon-14 beta particles (156 keV) substantially exceed the maximum tritium beta energy (18.6 keV). In addition, since the concentration of tritium (MBq/L) usually greatly exceeds the levels of carbon-14 (kBq/L), the tritium window (Channel A) is set to detect beta energies from 0 to 18.6 keV, and Channel B is set from 35 to 110 keV to detect carbon-14.

Current practice requires all radiation workers exposed to an annual intake of $^{14}CO_2$ that could exceed 5 mSv to participate in the urine bioassay program. Urine samples are routinely counted for only 0.5 minutes, a sufficient period for acceptably accurate measurement of tritium burden. However, the uncertainty in low level carbon-14 activities for this short count time is greater than required for acceptable accuracy, and a positive carbon-14 detection in the initial screen triggers a 10-minute recount to reduce counting error [6].

The use of linear interpolation to interpret intakes is not sufficiently accurate since the excretion rate function changes rapidly over the weeks after an intake of $^{14}CO_2$. A conservative estimate could be determined, assuming that an acute intake had occurred just following the previous sample. The approach used in ICRP-54 is to assume an intake at the mid-point of the sample period, but such an assumption can lead to bias. The OPG (Ontario Power Generation) applied a Most Probable Dose calculated on the basis of an equally-probable time of intake over the sample period [4]. Using this approach requires the acceptance of a small proportion of false positive results (below 5 %), most resulting in assigned whole body doses of a few 10's of μSv. Other approaches, such as the construction of an entirely separate bioassay program based on breath analysis, were considered; however, it was found to be much less cost effective, and to present problems of identifying and collecting all appropriate samples [6].

3.2 Dosimetry for Insoluble Carbon-14 Particulates

At OPG NPPs, extensive analyses of material removed from the annulus system characterized the forms of particulate containing carbon-14 both physically and chemically. The samples were also tested biologically by intratracheal instillation into the lungs of rats to measure clearance behavior. Both approaches indicated insoluble materials that cleared from the lungs with approximately ICRP Class Y characteristics [6]. Such material is eliminated from the body primarily in feces, not in urine.

A method was developed to detect carbon-14 in the lung directly using phoswich counting of the internal bresstrahlung photons since the interpretation of low levels of fecal excretion is uncertain and to provide positive assurance that large lung-burdens were not present and undetected due to very slow clearance [7]. Since these photons cover a spectrum of energy and this spectrum lies almost entirely below 30 keV, this developed method was necessarily somewhat insensitive.

4. Conclusion

Internal dosimetry is an often complex practice, of equivalent importance in some radiation programs to external dosimetry. Several foreign research institutes have performed the studies on the internal dosimetry for carbon-14 and make some progress. However, Korean nuclear facilities have not yet prepared clearly for the internal dosimetry for carbon-14. Thus, it is demanded to improve the capability to collect, analyze and interpret bioassay excretion samples and this is an essential element of the internal dosimetry program for carbon-14.

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