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The Status of Monitoring of Cabon-14 Emissions from LWRs in Korea

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1. Introduction

A pure beta emitter with a half-life of 5,730 years, carbon-14 (^{14}C) has a maximum energy of 156 keV and an average energy of 45 keV and is generated from natural factors such as cosmic rays and artificial factors such as nuclear experiments, reprocessing plants, and nuclear power plants (NPPs). The nuclear reactions that produce ^{14}C in reactors with thermal neutrons are $^{17}O(n,\alpha)^{14}C$, $^{14}N(n,p)^{14}C$.

When it is released in the form of $^{14}CO_2$ through the exhaust port of NPPs, the substance is retained in the bodies of plants and animals through plants' photosynthesis and the ingestion of plants. When plants containing ^{14}C are ingested as food, the substance is introduced into the human body and causes radiation exposure. Consequently, regulatory agencies require NPP operators to evaluate and to monitor the amount of ^{14}C emitted into the atmosphere through the exhaust port of NPPs. The present study examines the current state of regulation of ^{14}C in Korea and the United States, its effect on ^{14}C emissions, and the off-site dose calculation of local resident based on evaluations recently conducted at LWRs domestically.

2. Standards for ^{14}C emission

Most NPPs in the United States determine emissions of ^{14}C by using theoretically calculated values without separately collecting samples, and European NPPs evaluate and monitor ^{14}C emissions mainly by collecting samples. Japan does not monitor ^{14}C. The present section examines the current state of the monitoring of and regulations on ^{14}C emissions in Korea and the United States.

2.1 Domestic standards

As for Korean regulations on ^{14}C emitted from NPPs, emissions must not exceed the concentration limit within 10,000 Bq/m^3 (^{14}C dioxide form), stipulated by Article 6 (Effluent Concentration Limit) of the NSSC Notice 2013-49 on Standards for Radiation Protection, etc., and Article 16 (Prevention of Hazards to Environment) which stipulates the annual dose limit at the boundary of the exclusion area due to emissions in a gaseous state:

- Human organ equivalent dose by particle radioactive substances, ^{3}H, ^{14}C and radioiodine: 0.15 mSv

In addition, when operating multi-nuclear reactor facilities at one site, an annual effective dose limit of 0.25 mSv at the boundary of the exclusion area is stipulated. In addition, in accordance with NSSC Notice No. 2013-4 "Regulation on Survey of Radiation Environment and Assessment of Radiological Impact on Environment in Vicinity of Nuclear Power Utilization Facilities," ^{14}C must be periodically investigated in air and agricultural/livestock product samples from the environments surrounding NPPs, and the lowest limit of detection 0.25 Bq/g-C, is stipulated for environmental radioactivity analysis.

2.2 U.S. standards

According to 10 CFR Part 20 Appendix B of the United States, the control standard for release into the air is 11,100 Bq/m^3 (^{14}C dioxide form), a value similar to that of the release control standard in Korea. However, 10 CFR Part 50 Appendix I states, "The calculated annual total quantity of all radioactive iodine and radioactive material in particulate form above background to be released from each light-water-cooled nuclear power reactor in effluents to the atmosphere will not result in an estimated annual dose or dose commitment from such radioactive iodine and radioactive material in particulate form for any individual in an unrestricted area from all pathways of exposure in excess of 0.15 mSv to any organ" , so that while the human organ equivalent dose constraint of 0.15 mSv is identical to the Korean, unlike the latter, it does not specifically stipulate ^{3}H and ^{14}C.

In 2009, the NRC of the United States announced a revised version of the Regulatory Guide 1.21 (Measuring, evaluating, and reporting radioactive material in liquid and gaseous effluents and solid waste) and added contents related to ^{14}C. According to this revised version (Rev. 2), the radioactive effluents from commercial nuclear power plants over the same period have decreased to the point that ^{14}C is likely to be a principal radionuclide in gaseous effluents. Because the dose contribution of ^{14}C from liquid radioactive waste is much less than that contributed by gaseous radioactive waste, evaluation of ^{14}C in liquid radioactive waste is not required. Licensees should evaluate whether ^{14}C is a principal radionuclide for gaseous releases from their facility

Other exposure pathways are considered significant if a conservative evaluation yields an additional dose increment equal to or more than 10 percent of the total from all exposure pathways(Reg. Guide 1.109). The quantity of ^{14}C discharged can be estimated by sample measurements or by use of a normalized ^{14}C source term and scaling factors based on power generation or by use of the GALE code from NUREG-0017. Most U.S. NPPs evaluate ^{14}C emissions by using theoretically calculated values according to this guide.

3. Current state of ^{14}C emissions from Korean LWR

The present section reviews the results of the evaluation of emissions and the off-site dose calculation of local resident from the ^{14}C sampling and analysis recently conducted at pressurized light water reactors (LWRs) in Korea.

3.1 ^{14}C sampling and analysis

At LWRs in Korea, samples are periodically collected from the exhaust port of fuel buildings, auxiliary buildings, and radioactive waste buildings (or complex buildings), which make up the bulk of the channels that release ^{14}C, and are analyzed.

For the sample collection method, $^{14}CO_2$ is collected by using the bubbler of a NaOH solution, and the related chemical formula is as follows:

$$CO_2 + 2NaOH \rightarrow Na_2CO_3 + H_2$$

After the addition of a fluorescent liquid, the collected samples are analyzed using a liquid scintillation counter. Because tritium emits beta rays with a maximum energy of 18.6 keV and an average energy of 5.7 keV and ^{14}C emits beta rays with a maximum energy of 156 keV and an average energy of 45 keV, respectively, when conducting analysis using a liquid scintillation counter, measurements are made by setting the energy band at 20 keV-156 keV to minimize the effect of tritium(^3H).

3.2 Results of the evaluation of ^{14}C emissions

There are two methods for evaluating ^{14}C emissions, one for theoretically calculating the amount of ^{14}C generated, as stipulated in Regulatory Guide 1.21, and one which involves directly collecting samples from the exhaust port of NPPs and analyzing them. With the Korean OPR-1000 NPP as the standard, the present study compares in Table I the theoretically calculated emissions and the values actual measured from direct sampling.

The theoretical calculation method for obtaining the amount of ^{14}C generated is as follows:

$$^{14}C \text{ production} = N \cdot \sigma \cdot \Phi \cdot \lambda \cdot m \cdot t \qquad (1)$$

λ=decay constant(sec^{-1})

t=operation time(sec)

m=coolant mass(g)

N=concentration of nuclides(^{17}O, ^{14}N)(atoms/g)

σ=cross section(cm^2)

Φ=thermal flux($n/cm^2 \cdot sec$)

98% of the ^{14}C generated is in a gas form. IAEA TRS No. 421 reported $^{14}CO_2$ emission rates of 5-25% at American and European NPPs. Based on that report, the present study presupposed that 30% of the ^{14}C values determined here were emitted in the form of $^{14}CO_2$.

Table I: Comparison of ^{14}C emission values(calculation vs. sampling)

Periods	Calculated values[TBq]	Values from direct sampling [TBq]
2nd Quarter 2013	2.34E-02	2.62E-03
3rd Quarter 2013	2.34E-02	2.10E-03
4th Quarter 2013	2.34E-02	2.08E-03

a. Note: Data from one OPR-1000 Unit in 2013

According to the results in Table I, where comparisons are made based on cases of measurements at one OPR-1000 NPP, the emissions directly measured from collected samples were approximately 1/10 the level of emissions derived from theoretical calculations of the amount of ^{14}C generated. This disparity seems to stem from complex effects, including liquid emissions of ^{14}C, $^{14}CO_2$ emission rates, and measurement errors.

In addition, according to Table II, which compares the 2013 emission data and effluent control limits of the OPR-1000 NPP, the concentration of ^{14}C at the boundary of the exclusion area, considering atmospheric dispersion, was approximately 0.0002%~0.0003% of the effluent control limit and therefore adequately low.

Table II: Comparison of the 2013 emission data and effluent control limit(ECL)

Items	Unit 1	Unit 2
Annual ^{14}C emission value[Bq]	2.970E+10	3.824E+10
Concentration of ^{14}C at the boundary of the exclusion area [Bq/m^3]	2.021E-02	2.775E-02
Percentage of the ECL[%]	0.0002	0.00027

a. Note: Data from OPR-1000 NPP in 2013

3.3 Off-site dose calculation of local resident by ^{14}C

To grasp the effect of the residents' radiation exposure doses due to ^{14}C emissions, the present study shows in Table III the results of dose evaluation conducted in 2013 at the OPR-1000 NPP. In comparison with 0.15 mSv, (the reference value for organ equivalent doses presented in NSSC Notice No.

2013-49 Article 16 (Prevention of Hazards to Environment)), the organ equivalent doses due to ^{14}C fell within approximately 0.8% of the reference value, thus being low. While the organ equivalent dose results including all nuclides such as tritium likewise were adequately lower than the design limits, ^{14}C accounted for approximately 95% of the total organ equivalent dose.

Table III: Organ equivalent dose

Items	Unit 1	Unit 2
Organ equivalent dose by only ^{14}C [mSv]	1.158E-03	1.83E-03
Percentage of Limit[%]	0.772	0.789
Organ equivalent dose by particle radioactive substances, ^{3}H, ^{14}C and radioiodine [mSv]	1.222E-03	1.251E-03

a. Note: Data from OPR-1000 NPP in 2013

According to the results of the off-site dose calculation of local resident conducted in 2013 at a site equipped with six NPPs, including the OPR-1000 NPP used in the present study, the effective dose amounted to 1.203E-02 mSv. This was less than approximately 5% of the effective dose limit of 0.25 mSv at the boundary of exclusion area in cases where multi-nuclear reactor facilities are operated at one site, as stipulated in NSSC Notice No. 2013-49 Article 16 (Prevention of Hazards to Environment), and therefore adequately low. However, most of the contribution to the total effective dose was due to the effect of gases, and, in particular, the effect of ^{14}C amounted to approximately 91.61%, thus taking up the bulk, with the remainder due almost entirely to the effect of tritium.

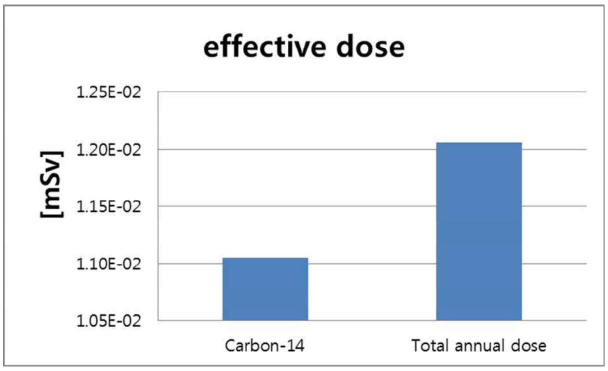

Fig. 1. Effective dose at a site equipped with six NPPs

4. Conclusions

In the present study, Korean and U.S. standards for ^{14}C emission monitoring were compared and the current state of emissions in Korea was examined. While most U.S. NPPs evaluate ^{14}C emissions based on theoretical calculations, Korean NPPs do so by directly collecting and measuring samples. When the results of the evaluation of ^{14}C emissions generated from the OPR-1000 NPP in Korea were recently reviewed, the limit stipulated in Article 6 (Effluent Concentration Limit) and Article 16 (Prevention of Hazards to Environment) of NSSC Notice No. 2013-49 "Standards on Radiation Protection, etc." was amply satisfied. However, because 90% or more of the effect on the residents' radiation exposure doses was demonstrated to consist of gaseous ^{14}C effluents, to minimize the emission of ^{14}C gas, NPP operators must actively study and practice ways of reducing the release of ^{14}C gas, such as periodically conducting gas stripper operation and the release of ^{14}C in a liquid form.

REFERENCES

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