The Status of Monitoring of Carbon-14 Emissions from Nuclear Power Plants in Korea – 15188

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ABSTRACT
In the present study, we try to configure C-14 generation and the release of C-14. From the average release of CO2 measurement, averaged is 6.12%, which is still in the range of EPRI proxy generation rate for effluent release without recombiner. 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 14C amounted to approximately 91.61%, thus taking up the bulk, with the remainder due almost entirely to the effect of tritium. Because 90% or more of the effect on the residents' radiation exposure doses was demonstrated to consist of gaseous 14C effluents, to minimize the emission of 14C gas, NPP operators must actively study and practice ways of reducing the release of 14C gas, such as periodically conducting gas stripper operation and the release of 14C in a liquid form.

INTRODUCTION
A pure beta emitter with a half-life of 5,730 years, carbon-14 (14C) 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 14C in reactors with thermal neutrons are 17O(n,α)14C, 14N(n,p)14C.
When it is released in the form of 14CO2 through the exhaust port of NPPs, the substance is retained in the bodies of plants through plants' photosynthesis and animals by the ingestion of plants. When plants containing 14C 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 14C emitted into the atmosphere through the exhaust port of NPPs. The present study examines the current state of regulation of 14C in Korea and the United States, its effect on 14C emissions, and the off-site dose calculation of local resident based on evaluations recently conducted at LWRs domestically. This present study examines C-14 generation and released from OPR1000 reactor and identifies form of release from many OPR reactors. EPRI has conclude that the C-14 generation will be different for each process, however the C-14 form comparison will be proportional for type of reactor.

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

Korean standards
As for Korean regulations on 14C emitted from NPPs, emissions must not exceed the concentration limit within 10,000 Bq/m^3 (14C 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, 3H, 14C 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," 14C 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.

USA 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 (14C 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 3H and 14C.
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 14C. 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 14C is likely to be a principal radionuclide in gaseous effluents. Because the dose contribution of 14C from liquid radioactive waste is much less than that contributed by gaseous radioactive waste, evaluation of 14C in liquid radioactive waste is not required. Licensees should evaluate whether 14C 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 14C discharged can be estimated by sample measurements or by use of a normalized 14C source term and scaling factors based on power generation or by use of the GALE code from NUREG-0017. Most U.S. NPPs evaluate 14C emissions by using theoretically calculated values according to this guide.

CURRENT STATE OF CARBON-14 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 14C sampling and analysis recently conducted at pressurized light water reactors (LWRs) in Korea.

14C sampling and analysis
According 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 14C, and are analyzed.
For the sample collection method, 14CO2 is collected by using the bubbler of a NaOH solution, and the related chemical formula is as follows:

$CO_2 + 2NaOH \rightarrow Na_2CO_3 + H_2$

$Na_2CO_3 + 2NH_4Cl + 2H_2O \rightarrow (NH_4)_2CO_3 + 2NaCl$

After the addition of a fluorescent(Ultima Gold XR) 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 14C 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).

Results of the evaluation of 14C emissions

According There are two methods for evaluating 14C emissions, one for theoretically calculating the amount of 14C 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 14C generated is as follows:

$$14C\ production = N \cdot \sigma \cdot \Phi \cdot \lambda \cdot m \cdot t \quad (1)$$

λ=decay constant(sec^{-1})

t=operation time(sec)

m=coolant mass(g)

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

σ=cross section(cm^2)

Φ=thermal flux($n/cm^2 \cdot sec$)

98% of the 14C generated is in a gas form. IAEA TRS No. 421 reported 14CO2 emission rates of 5-25% at American and European NPPs. Based on that report, the present study presupposed that 30% of the 14C values determined here were emitted in the form of 14CO2.

Table I: Comparison of 14C 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 14C generated. This disparity seems to stem from complex effects, including 14C in liquid (e.g. liquid emissions, wet solid waste from treatment of liquids), 14CO2 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 14C 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 14C emission value[Bq]	2.970E+10	3.824E+10
Concentration of 14C 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

Carbon-14 release form

Table III reflect C-14 generation based on calculation based on eq.(1) and measurement C-14 release in the form of CO2 from released pathways (exhaust port of fuel buildings, auxiliary buildings, and radioactive waste building (or complex buildings).

Table III: Percentage of CO_2 release from generation

Reactor/Periods	Production(Bq)	CO_2 release (Bq)	CO_2 release(%)
Hanul 3			
2013 Q2	2.29E+10	2.62E+09	1.12E-01
2013 Q3	2.29E+10	2.10E+09	8.97E-02
2013 Q4	2.29E+10	2.08E+09	8.89E-02
Average			9.69
Hanbit 5			
2013 Q2	2.56E+10	2.12E+09	8.28E-02
2013 Q3	2.56E+10	3.32E+07	1.30E-03
2013 Q4	2.56E+10	2.43E+09	9.49E-02
Average			5.97
Hanbit 6			
2013 Q2	2.56E+10	3.97E+08	1.55E-02
2013 Q3	2.56E+10	1.87E+08	7.30E-03
2013 Q4	2.56E+10	1.49E+09	5.82E-02
Average			2.70
Total Average			6.12

EPRI has make a proxy generation of C-14 in the reactor based on calculation of C-14 generation and measurement of C-14 released from many measurement. Figure 1 reflects proxy generation for OPR1000 from Hanul3, Hanbit5 and Hanbit 6. Averaged generated C-14 is 2.49E10 Bq, the average release of CO_2 is 6.12%. Based on EPRI study, for gas effluent processing system without recombiner, the chemical form of release will be 5-30% CO_2 , which is still in the range of EPRI proxy generation rate. Without recombiner most of the gaseous release will be assumed to be CO_2.

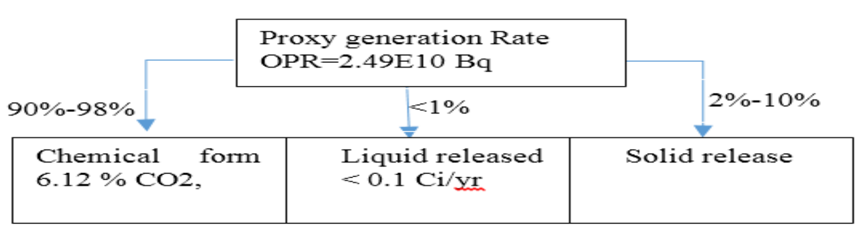

Figure 1. Proxy generation rate for OPR1000 in Korea

OFF-SITE DOSE CALCULATION OF LOCAL RESIDENT BY CARBON-14

To grasp the effect of the residents' radiation exposure doses due to 14C emissions, the present study shows in Table IV 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 14C 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, 14C 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 14C [mSv]	1.158E-03	1.83E-03
Percentage of Limit[%]	0.772	0.789
Organ equivalent dose by particle radioactive substances, 3H, 14C 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 14C amounted to approximately 91.61%, thus taking up the bulk, with the remainder due almost entirely to the effect of tritium.

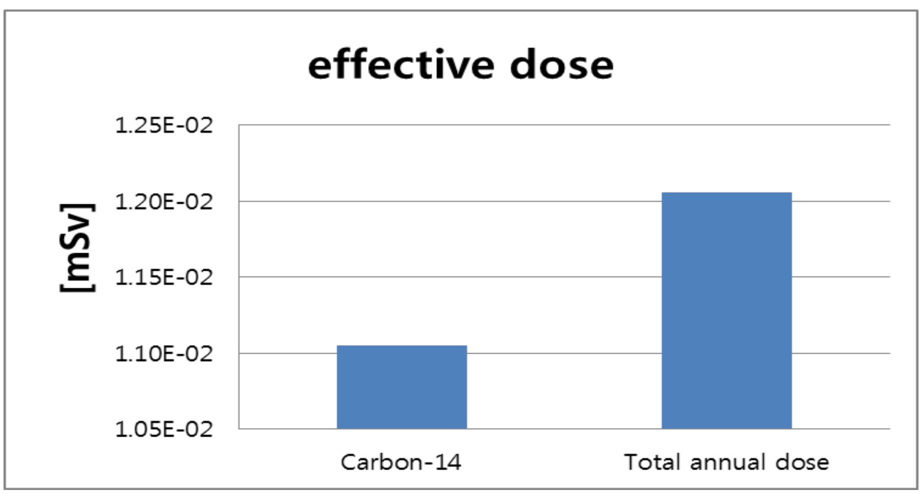

Figure 2. Effective dose at a site equipped with six NPPs

CONCLUSIONS

In the present study, Korean and U.S. standards for 14C emission monitoring were compared and the current state of emissions in Korea was examined. While most U.S. NPPs evaluate 14C emissions based on theoretical calculations, Korean NPPs do so by directly collecting and measuring samples. When the results of the evaluation of 14C 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. From the average release of CO2 measurement, averaged is 6.12%, which is still in the range of EPRI proxy generation rate for effluent release without recombiner. However, because 90% or more of the effect on the residents’ radiation exposure doses was demonstrated to consist of gaseous 14C effluents, to minimize the emission of 14C gas, NPP operators must actively study and practice ways of reducing the release of 14C gas, such as periodically conducting gas stripper operation and the release of 14C in a liquid form.

ACKNOWLEDGMENT

This work was supported by the Nuclear Safety Research Program through the Korea Radiation Safety Foundation (KORSAFe), granted financial resource from the Nuclear Safety and Security Commission (NSSC), Republic of Korea (No. 1305004).

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