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Study on C-14 Generation and Release in Pressurized Water Reactor

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1. Introduction

During nuclear reactor operation, radionuclides are produced and released in the form of gas effluent or liquid release. Regulatory body required that radioactive material released from nuclear power plant is as low as reasonably achievable. The quantities release depend on type of reactor and radioactive waste management incorporate to the reactor. The quantity expected to be released is called source term. This quantitates set forth isotope of listing released. This is very useful for regulatory body to review release from the reactor to confirm that dose to individual will meet ALARA criterion [1].

One of the list radionuclide released is Carbon 14 (C-14) with 5370 years of half-life and pure beta emitter with maximum energy 156 keV, and average energy of 45 keV. As improvement in nuclear power plant effluent management practices, C-14 is considered to be the principal radionuclide giving main dose contribution. Moreover, C-14 in the form of CO_2 is environmental mobility in nature and ease of assimilation into living matter.

EPRI (Electric Power Research Institute) study concludes that the C-14 generation will be different for each process; however the C-14 form comparison will be proportional for type of reactor [2]. This present study examines C-14 generation and released from OPR1000 reactor and describes the release form from OPR1000 reactors.

2. Methods and Results

We study on the generation and release of C-14 in the PWR reactor and compare it with some study done by EPRI.

2.1 Carbon-14 generation

C-14 radionuclide can be generated naturally through cosmic or artificially inside reactor operation or utilization in the medical or industrial application. During nuclear reactor operation, C-14 is generated in the fuel, in core structure material, and in the cooling water. C-14 generation in the fuel and core structure material is considered very low to be release in normal operation. This is because C-14 generated in fuel is still contained inside matrix fuel and cladding, while C-14 generated in the core structure is still retained in within the metal. This kind of C-14 generation will be important during reprocessing fuel or disposal (decommissioning) nuclear reactor. The most C-14 generation to be released comes from primary water coolant reactor. The main radionuclide contributed to generation C-14 is O-17 and dissolved N-14. This is because O-17 and N-14 have high thermal neutron capture cross section.

To estimate C-14 generated in reactor water coolant, we can do by calculation with considering number of target (O-17 or N-17), effective cross section, and flux neutron, with equation:

$$A= N_{t.} \sum(\sigma_i \ \varphi_i) .\lambda. m .t \quad (1)$$

A = activity of the generated C-14 per sec
N_t = number of target (atom /g of coolant)
σ_i = cross section (cm^{-2})
φ_i = thermal flux ($n/cm^2.sec$)
λ = decay constant (sec^{-1})
m = mass of coolant (kg)
t = operation time (s)

Cycle of fuel and all energy of neutron (thermal, intermediate and fast) should be considered to make the calculation more realistic.

2.2 Carbon-14 release and pathways

The C-14 generation mainly released from generation in the coolant water. Gaseous releases constitute the main release path from this source. In PWR, C-14 released from waste gas system is in the mix form between organic carbon and carbon dioxide. Compare to BWR, C-14 is released mainly as carbon dioxide in gaseous waste, this is because BWR coolant system is in oxidizing environment. Some of the release also in the form of liquid waste, which is very less compare to gaseous release so that giving less dose contribution compare to gas effluent [3]. Some of C-14 generated is in the form of solid resin, which will important while in later disposal.

The pathways of gas release for PWR is considered to be from: waste gas system, containment vent, containment purge, auxiliary building vent, and fuel handling building. Waste gas system receives C-14 from stripping of primary coolant during normal operation and also from degassing the primary coolant (cold-shutdown). There is also some C-14 that leakage from primary coolant system and will enter the containment and auxiliary building.

2.3. Measurement C-14 sample and analysis

In Korea, gas effluent releases are collected periodically for measurement release. Sample are periodically collected from exhaust port of fuel buildings, auxiliary buildings, and radioactive waste building (or complex buildings), which make up the bulk of channels that release C-14, and analyzed. Sample collection method, $^{14}CO_2$ is collected by using the bubbler of a NaOH solution, and following reaction:

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

After the addition of fluorescent liquid, the collected sample is analyzed using 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 maximum energy of 156 keV and an average energy of 45 keV, respectively, when conducting analysis using a liquid scintillator counter, measurements are made by setting the energy band at 20 keV-156 keV to minimize the effect of tritium (^3H).

2.4. Result

Table 1 reflects C-14 generation based on calculation following eq.(1) and measurement C-14 in the form of CO_2 from released pathways (waste gas system, containment vent, exhaust port of fuel buildings, auxiliary buildings, and radioactive waste building or complex buildings).

Table 1: Percentage of CO_2 release from generation

Reactor/ Periods	Calculated (Bq)	Measurement CO_2 release (Bq)	percentage 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

The result of percentage C-14 in the form of CO_2 is on average 6.12 % from the total calculated C-14 generated in the reactor coolant system. In TRS No. 421, C-14 released in the form of CO_2 for US and Europe PWR reactor is around 5-25% [4]. EPRI study also confirmed that C-14 in the form of CO_2 estimated around 5-30% and the remainder is organic form. The smaller result for inorganic form than that of organic form is caused by recombiner system is not used in for gas processing system. While recombiner system is used in gas processing system, C-14 to be released in gas form is assumed to be CO_2.[5]

EPRI has made proxy generation rate to describe C-14 released from specific type of reactor. Picture 1 reflects modified of proxy generation for OPR1000 from Hanul 3, Hanbit 5 and Hanbit 6. Averaged generation C-14 from these reactors is 2.49E10 Bq, and the averaged release of C-14 in the form CO_2 is about 6.12%.

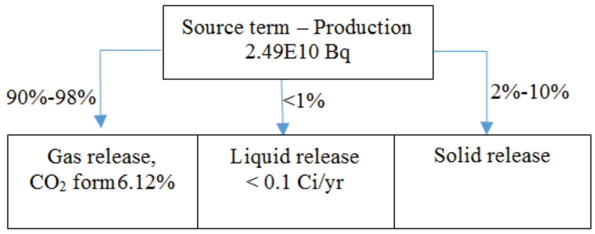

Fig.1. Proxy generation rate for OPR1000

3. Conclusions

The result of this work confirms the results of EPRI report with respect to generation, chemical form, and release of C-14 from PWR reactor. EPRI also confirmed that C-14 produced in PWR coolant system is mainly in methane form as result of PWR operation in reducing environment. However, small fraction of C-14 can be released in the form CO_2. The percentage release of C-14 in the form CO_2 from Hanul 3, Hanul 4, and Hanbit 6 is averaged 6.12%. This result also confirms EPRI's proxy generation rate that C-14 in the form of CO_2 is about 5-30% for gas processing system without recombiner.

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