Production and Release of Carbon-14 at Younggwang unit-4

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1. Introduction

In PWR, Carbon-14 is produced as activation product in the coolant, fuel and structural material during normal operation of nuclear power plant [1]. Carbon-14 has a radioactive half-life of 5730 yr and, when released to the atmosphere, becomes a long term contaminant to environment. Since the amount of Carbon-14 released from PWR is small and it is not easy to detect, it is not selected as a main monitoring nuclide for environmental release in PWR in general.

Korean regulatory authority (KINS) raised the issues on a practical evaluation of Carbon-14 including released amount, environmental behavior and dose assessment. Therefore, Korea Hydro and Nuclear Power Company (KHNP) has performed the monitoring for Carbon-14 at Yonggwang-4 unit since January 2003. The samples are collected and analyzed monthly.

2. Method and Results

2.1 Production of Carbon-14 at PWR

The most important production reactions for 14C formation in PWRs are $^{17}$O(n, α)$^{14}$C and $^{17}$N(n, p)$^{14}$C. The natural isotopic abundance of $^{17}$O is 0.038% of oxygen, which is a major constituent of the coolant water and oxide fuel. Production of $^{14}$C form $^{17}$O is therefore unavoidable. The isotopic abundance of $^{14}$N is 99.62% of N, which is present as an impurity. Production of $^{14}$C from the activation of $^{14}$N is difficult to estimate because the N concentrations in the coolant water are not well-known [4]. Generally, during normal operation of PWR, nitrogen addition results from dissolved gas in the reactor makeup water that is charged to the primary system to compensate for system leakage and the reactor coolant bleed required for boron dilution. The production of $^{14}$C within the coolant water can be calculated by using the following equation

$$ A = N_0 \cdot \sigma \cdot \phi \cdot m \cdot t \cdot CF \cdot S \, \text{(Ci/cycle)} $$

Where:

- $N_0$ = atom concentration in the reactor coolant water (atoms/kg H$_2$O)
- $\sigma$ = effective thermal cross section (cm$^2$)
- $\phi$ = thermal neutron flux, 6.62E+13 n/c m$^2$-s
- $m$ = mass of core water, 1.6E+04 kg
- $t$ = conversion factor, (sec/cycle)
- $CF$ = plant capacity factor
- $S$ = 1.037E-22 Ci/atom

For $^{14}$C production from $^{17}$O activation, $N_0 = 1.27E+22$ atoms $^{17}$O/kg(H$_2$O) and effective thermal cross section $\sigma = 1.48E-25$ cm$^2$ are used in the above equation. The production rate is 8.49 curies/cycle. For $^{14}$C production from $^{14}$N activation $N_0 = 1.28E+20$ atoms $^{14}$N/kg (H$_2$O) and $\sigma = 1.14E-24$ cm$^2$ are used in the above equation. The CF value is an average capacity factor 0.87. The production rate is 0.66 curies/cycle. The production of 14C from these sources during the fuel cycle is 9.15 curies.

2.2 Trapping and Sample Treatment of Carbon-14

In order to monitor C-14 in PWR, KHNP devised C-14 sampling apparatus which can collect CO$_2$ and Hydrocarbon separately. It is composed of three main parts, that is, primary CO$_2$ sampler, a Hydrocarbon oxidization assembly and a secondary CO$_2$ sampler. The primary CO$_2$ sampler has one water bubbler and two NaOH bubblers. Then the Hydrocarbon oxidization assembly which is consisted of furnace and catalyst column converts organic $^{14}$C into CO$_2$. The catalyst is composed of equal mixture of Alumina with Palladium coating (1.0%) and Platinum coating (0.5%).
To trap $^{14}$C, we used 2M NaOH solution (trapping solution) from the air added the NH$_4$Cl to prevent the precipitation of Ca(OH)$_2$, because the Ca(OH)$_2$ is precipitated together the CaCO$_3$, we can’t separate them because of the same color and the same form. So we add the CaCl$_2$ to precipitate a CaCO$_3$. And we drop the HNO$_3$(nitric acid) to regenerate the CO$_2$ from the CaCO$_3$, and then the absorber catches the generated CO$_2$. After mixing it with the scintillator, the C-14 is measured by liquid scintillation counter.

2.3 Results of Monitoring for Carbon-14

We had monitored carbon-14 at Yonggwang unit 4 since from January-2003 to December-2004. According to the analysis results, total activity was estimated to be about 0.147TBq/GWe-yr which was about 67% of world’s PWR average(0.22TBq/GWe (UNSCEAR 2000)).

3. Conclusion

The production of $^{14}$C was calculated about 6.5 Ci/yr. While the release amount is 3.97Ci/yr in younggwang unit-4. Our future works is that these samplers will be installed additionally to collect several kinds of carbon-14 at the other PWR plants(CE, WH, FRAMATOM types). Based on the results, we will evaluate the environmental effects of carbon-14 at PWR plants.

REFERENCES

1. N. Sion, Detection and Measurement of C-14 in Nuclear Power Plants Gaseous Effluents, Ontario Power Generation, Toronto

