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Estimation of Amounts of Carbon-14 Formation in and Release from HANARO

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1. Introduction

Carbon-14 (half-life 5,730 years; beta decay to N-14 with max. energy of 156 keV, average 49 keV) is one of the potential sources causing the internal radiation exposure. The release of C-14 to the environment is regulated by a Notice of the MOST on the radiation protection.[1] The C-14 activity in the exhaust air in the form of CO and CO_2 is limited to 0.1 MBq/m^3. Meanwhile, C-14 is one of the elements categorized as a group of gaseous effluents together with radioactive particulates, tritium, and radioactive iodine. The equivalent dose rate to organs from this group is limited to 0.15 mSv a year.

There have been long discussions, unconcluded yet, on whether we have to monitor and control the release of C-14 from nuclear facilities. Even if the amount of C-14 released from the PWR's has not been proven negligible, a report on the C-14 monitoring techniques indicated that the environmental monitoring at a site of heavy water power reactor is more important than that at a PWR site.[2] The major source of C-14 in a heavy water reactor is the $^{17}O(n,\alpha)$ reaction in the heavy water. Because the HANARO research reactor uses quite amount of heavy water as the reflector-moderator, the C-14 formation in the HANARO takes attention, too.

In this paper, we present the results of neutronics calculation on the C-14 formation rate in the HANARO. We also present the estimation of the release to the environment which is based on crude, but conservative assumptions.

2. Estimation of the Amount of C-14 Formation

2.1 Sources of C-14 Formation

C-14 is formed as a product of nuclear reactions such as $^{14}N(n,p)$, $^{17}O(n,\alpha)$, $^{13}C(n,\gamma)$, and $^{16}O(n,\gamma)$ that are dominant in the thermal energy region, and $^{15}N(n,d)$ and $^{16}O(n,^3He)$ reactions in the fast region. The first two reactions are the ones we took into account in this estimation. Any formation by the other reactions was neglected because of small content of reactants, small cross section, and/or small neutron flux at a region of interest.[3] Contrarily, the cross sections of the first two reactions are as fairly large as the order of barns and the contents of N-14 and O-17 are relatively large in HANARO.

In HANARO, oxygen exists in the light water as the coolant and pool water and exists in the heavy water in the reflector tank. The abundance of O-17 in heavy water is known as 0.055%,[2] which is much higher than the natural abundance of 0.038%. Oxygen also exists in structural materials: Zircaloy-4, the material of the reflector tank and the flow tubes, contains oxygen of 1,400 ppm and Hf as the neutron absorber contains up to 400 ppm of oxygen. Unlikely to the UO_2 fuel of power reactors, the uranium silicide fuel of HANARO does not contain oxygen.

Nitrogen in the air is dissolved in the water at the solubility of 2 mg of N_2 in 100 g of water at room temperature. Particularly in HANARO, the nitrogen gas is used as the shooting gas for sample in and out for the Neutron Activation Analysis (NAA), thus the formation of C-14 in three NAA holes should be taken into account. Meanwhile, nitrogen is one of the impurities of structural materials, up to 80 ppm in zircaloy-4. The natural abundance of N-14 is 99.6%.

2.2 Estimation of Formation Rate

We used a simple formula to calculate the C-14 formation rate from either $^{14}N(n,p)$ or $^{17}O(n,\alpha)$ reaction. Recall that both reactions are thermal reactions. The formation rate R in a region is calculated as

$$R = \phi_{th} N \sigma_M , \qquad (1)$$

where ϕ_{th} is the thermal neutron flux averaged over a region of interest and N is the total number of atoms of a reactant (N-14 or O-17) in that region. σ_M is the cross section averaged over the Maxwellian spectrum at 300 K; we used 1.827 b for $^{14}N(n,p)$ reaction and 0.235 b for $^{17}O(n,\alpha)$ that were processed from the ENDF/ B-VI.

We divided the whole HANARO into five regions not on a geometrical but on a material basis. Those are the heavy water reflector region, core region filled with light water, vertical irradiation holes filled with light water, NAA holes, and the reactor structure region that includes the structural materials in the reactor core and the inner shell of the reflector tank.

Table 1 shows the thermal neutron fluxes and some basic data for calculating the numbers of atoms. The thermal fluxes in all regions except the reflector region are the thermal group fluxes, of which upper energy is 0.625 eV, taken from the VENTURE calculation.[4] These values were very conservatively taken because of a difficulty in obtaining values averaged over the whole region; for instance, the flux in the core region is actually the flux at the central thimble of the core. For the reflector region, however, reflecting its importance in the total C-14 formation, we calculated the average flux rather realistically using the MCNP code.

Table 1. Raw Data for the Calculation of C-14 Formation Rate

	Reflector	Core	Irradiation holes	NAA holes	Structures
Source material of C-14 formation	heavy water	light water filling CT & IRs	light water filling irr. holes	N_2 shooting gas	Zr-4 and Hf in the core region
Mass of medium containing oxygen	3,440 kg of D_2O	237 kg of H_2O	226 kg of H_2O	-	524 kg of Zr-4, 37.6 kg of Hf
Mass of nitrogen contained (g)	68.8	4.7	4.3	12.7	41.9 in Zr-4
Thermal n. flux (1E+14 /cm^2·s)	0.665 (average)	3.06 (at CT)	1.39 (at IP15)	1.19 (at NAA3)	3.06

Table 2. C-14 Formation Rate

	Reflector	Core	Irrad. holes	NAA holes	Structures	Sum
Formation rate, 1E+14 #/sec						
from $^{17}O(n,\alpha)$	8.91	2.17	0.94	-	0.01	12.0
from $^{14}N(n,p)$	3.58	1.14	0.47	1.19	10.05	16.4
Sum, 1E+14 #/sec	12.5	3.31	1.41	1.19	10.06	28.4
TBq/year	0.121	0.032	0.014	0.012	0.097	0.28
Fraction (%)	44	12	5	4	35	

As shown in Table 2, the C-14 formation rate in HANARO was calculated as 0.28 TBq/year or, of equivalent, 7.4 Ci/year. Note that the availability of HANARO was assumed as 80% at 30 MW and the decay of C-14 was neglected due to its long half-life. This formation rate is very close to the estimation of ~28 TBq/GW_e·year in the CANDU-6 reactor[2] that is converted roughly to 0.28 TBq/year for 30 MW_{th}. Still, in a base of total amount, the formation in HANARO is estimated as only ~1/60 of the amount formed in one CANDU-6 reactor.

On the other hand, Table 2 also shows that the most important sources of C-14 in HANARO are the heavy water and then the structural material zircaloy-4. This finding is qualitatively consistent with previous studies summarized in Ref. 2: the major source of C-14 in CANDU reactor is the $^{17}O(n,\alpha)$ in the heavy water while half of the C-14 in a light water reactor is formed in structural materials.

3. Estimation of the Amount of C-14 Release

C-14 dissolved in the water exists mostly in the form of bicarbonate ion. The efficiency of the ion exchanger is known as 91~95%[2] in removing the bicarbonate ions. For the C-14 release rate calculation, we made several crude assumptions such as i) the efficiency of the ion exchanger is 90%, ii) all the C-14 not adsorbed in the ion exchanger is released to the atmosphere, iii) all the C-14 formed in the structural materials is escaped to and dissolved in the light or heavy water, and iv) all the C-14 formed in the NAA shooting gas is released to the atmosphere.

With the assumptions above, the rate of C-14 release to the atmosphere was calculated as 0.039 TBq/year. The exhaust rate of air from the HANARO reactor hall and reactor concrete island is 36,600 m^3/h, thus this amount is equivalent to 0.1 kBq/m^3 in average, which is only 0.1% of the C-14 exhaust management criterion, 0.1 MBq/m^3, in the MOST Notice. Meanwhile, the amount of 0.039 TBq/year is about 5~35% of airborne C-14 activities measured in CANDU sites in Canada and Wolsong in 1990's.[2]

4. Conclusion

In spite of very conservative assumptions made for the calculations, the amounts of C-14 formation in and release from HANARO were estimated as small enough. The C-14 activity in the exhaust air is only about 0.1% of the exhaust criterion given in the MOST Notice. The amounts are also as small as few percents in formation and few to several tens of percents of the amounts in a typical CANDU reactor. If we knew and applied rather realistic mechanism of the release to the environment, the estimated amount of release would decrease a lot.

In conclusion, because of so small amount of C-14 formation in and release from the HANARO reactor, it seems we do not need a monitoring system dedicated to C-14.

REFERENCES

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