The Effect of ^{3}He on the Tritium Generation in the CANDU PHWR

Bum-Jin Chung, Sin Kim and Min Chan Kim[*]

Department of Nuclear and Energy Engineering

Department of Chemical Engineering and Clean Technology Cheju National University, Jeju 690-756, Korea

ABSTRACT

Much tritium is generated in the CANDU PHWR, compared to the LWR, due to the neutron capture reaction of deuterium in heavy water. ^{3}He, the daughter nuclide of tritium, re-transforms to tritium with the cross-section of about 10^7 times as large as that of deuterium. A calculation model is proposed, which includes the ^{3}He contribution in the the tritium generation. The contribution of ^{3}He to the tritium generation in Wolsong NPP #1 was evaluated and compared to the measured data. This study concludes that the contribution of ^{3}He in coolant is very large but that in moderator is negligible due to the low solubility and ^{4}He cover gas.

Key words : *Tritium, Helium, Solubility, CANDU, PHWR*

1. INTRODUCTION

Tritium(^{3}H, denoted as T in this paper), the only radioactive isotope of hydrogen, decays to ^{3}He by the emission of a β^- particle with a half-life of 12.34 years. The amount of tritium in nature is about 70~140 MCi and annual increase is about 4~8 MCi. Until the early sixties, the increase of tritium in nature was largely due to nuclear weapon tests. However it shifted to the nuclear power plants according to the growing nuclear power generation(Sinclair, 1987).

Tritium is generated by the reactions of atmospheric atoms with cosmic rays, nuclear fission, and activation reactions such as ^{2}H(n, γ)T, ^{3}He(n, p)T, ^{6}Li(n, α)T, ^{7}Li(n, n α)T, ^{9}Be(n, α)^{6}Li, ^{10}Be(n, α)^{7}Li, ^{10}Be(n, 2 α)T, ^{11}B(n, n2 α)T. In LWRs(Light Water Reactors), tritium is mainly generated by ternary fission in the nuclear fuel and by neutron reactions with light elements such as boron and lithium in control rods or burnable poison dissolved in the primary water coolant. On the other hand, the amount of tritium generated in PHWR(Pressurized Heavy Water Reactor) by neutron capture reactions of deuterium in heavy water exceeds that in LWR by almost a 100 times(Johnson et al., 1992 ; Peterson and Baker, 1985). The annual amount of tritium generation in PHWR is 2,400 Ci/MWe and the amount of gaseous and liquid effluents of tritium in CANDU PHWR is more than 100 times as large as those in LWR.

Meanwhile in the reactor core, the ^{3}He which is the daughter nuclide of tritium is transformed to tritium again by neutron capture reaction. The existing calculation models on tritium generation neglect the contribution of ^{3}He due to its low solubility(KEPRI, 1989). However the neutron capture cross-section of ^{3}He is 1.6×10^7 times as large as that of deuterium. That means that even only the dissolved amount of 0.03 ppm of ^{3}He in heavy water is enough to produce the same amount of tritium as that produced by the deuterium of total heavy water. Furthermore, according to the Henry's law, the solubility of helium increases as the operating pressure increases.

This study dealt with the contribution of ^{3}He to the tritium generation. The solubilities of ^{3}He in heavy water at each operating condition, were evaluated and the heavy water operating systems which affect the solubility of ^{3}He were reviewed. As a sample case, this method was applied to Wolsong #1 (CANDU-6 PHWR).

2. SOLUBILITY OF ^{3}He IN HEAVY WATER

It is a generalized theory of gas solubility that as the solvent temperature is reduced and the pressure is increased, a gas is more soluble(Markham and Smith, 1995). The solubility of ^{3}He in heavy water depends on the operating conditions and the system configurations. The operating conditions of both the moderator and coolant in CANDU PHWR are tabulated in Table 1.

Table 1. The Operating Conditions of Moderator and Coolant in CANDU PHWR

Operating Conditions	Moderator	Coolant
Quantity	264 ton	240 ton
Temperature	70 °C	310 °C
Pressure	101.325 Pa	11.1 MPa
Flow rate	940 l/s	94520 l/s

A correlation for ^{3}He solubility in heavy water does not exist but there are some available correlations on ^{4}He solubility in light water. Apparently the solvent characteristics of light water and heavy water must be different and the solubility can be affected by gaseous and liquid impurities. However considering the state of the art of the correlations, we assume that the solubility of ^{4}He can be regarded as the solubility of ^{3}He since chemical characteristics of substances depend on the atomic number of substances.

Using the above assumption and the R. Battino's correlation, an experimental equation for the solubility of ^{4}He in light water at 101.325Pa (1 atm), the solubility of ^{3}He in heavy water was estimated to be 1.7 ppm(IUPAC, 1979) :

$$\ln X_l = -41.4611 + 42.5962 \left(\frac{T}{100}\right)^{-1}$$
$$+ 14.0094 \cdot \ln\left(\frac{T}{100}\right) \qquad (1)$$

where

X_l : the mole fraction solubility of helium at

    101.325Pa(1 atm) and

T : temperature in Kelvin.

Meanwhile the moderator and auxiliary systems should be considered. The moderator system is essentially a closed heavy water recirculating loop which serves to cool the heavy water moderator. The moderator auxiliary systems comprises the cover gas system, the moderator heavy water collection system, the moderator sampling system, the moderator liquid poison system, and the moderator purification system. The moderator cover gas system provides a controlled inert gas atmosphere over the free surface of the moderator in the calandria extension to prevent accumulation of deuterium and oxygen gases generated by radiolysis of the heavy water moderator(KEPCO, 1976). Considering the helium cover gas system, we can estimate that the ^{4}He is saturated in the moderator at the initial stage and very little of the ^{3}He produced by β^- decay of tritium dissolves. Therefore the ^{3}He contribution in the moderator to tritium generation could be negligible.

As seen in Table 1, the operating conditions of the coolant are about 583.7K(310°C) and 11.1MPa(KEPCO, 1976). In order to use the experimental results of Wiebe and Gaddy, the operating temperature of the coolant is assumed to be 588.7K[8]. Wiebe and Gaddy measured high pressure helium solubility in water at three different temperatures. From the experiments of Wiebe and Gaddy, the solubility of helium at 588.7K and 1.379MPa is 310.6ppm. Applying Henry's law, we can estimate the helium solubility at the actual operating conditions is approximately 1,180ppm.

$$\frac{n_g}{n_g + n_l} = 1.3956 \times 10^{-3} \quad \text{and}$$

$$\frac{w_g}{w_l} = 0.3106 \ \frac{g - ^4He}{kg - H_2O} \qquad (2)$$

where

n_g, n_l : the mole number of helium and water, respectively, and

w_g, w_l : the molecular weight of helium and water, respectively.

The heat transport systems comprise the coolant system and the associated auxiliary systems. The coolant system circulates pressurized heavy water through the reactor fuel channels to remove heat produced by fission of the natural uranium fuel. The associated auxiliary systems include the pressure inventory control system, the purification system, and the heavy water collection system(KEPCO, 1976). The purification system functions during normal reactor operation to limit activity and corrosion product buildup in the coolant by removing soluble and insoluble impurities. Hydrogen is added via this system to suppress oxygen generated from radiolysis of the heavy water. ^{3}He has not been yet controlled by the purification system because its quantities are negligible. However, as the solubility of ^{3}He in the coolant operating conditions is much larger than 0.03ppm, the contribution of ^{3}He to tritium generation may not be negligible. In other words, ^{3}He dissolved in the coolant, compared to that in the moderator, is expected to make a larger contribution to the tritium generation due to its operating conditions and system configurations.

3. CALCULATION OF THE TRITIUM CONCENTRATION

The tritium concentration in the coolant and the moderator as a function of plant operating time are calculated using the following assumptions:

1) There is no mixing between the coolant and the moderator.

2) The unrecoverable heavy water losses are made up with virgin heavy water.

3) The tritium concentration in replacement heavy water can be negligible.

4) The unrecoverable moderator losses are very small relative to those of coolant.

5) The total heavy water in the coolant and the moderator is constant.

6) The plant capacity factor is 80%.

The change rates of the atomic number of tritium and ^{3}He in heavy water can be given by:

$$\frac{d}{dt}(M \cdot N_T) = F \cdot N_F + \sum_{i=1}^{47}\phi_i \cdot \sigma_D^i \cdot N_D \cdot m \cdot a$$
$$+ \sum_{i=1}^{47}\phi_i \cdot \sigma_{He}^i \cdot N_{He} \cdot m \cdot a$$
$$- \lambda_T \cdot M \cdot N_T - L \cdot N_T$$
$$\frac{d}{dt}(M \cdot N_{He}) = \lambda_T \cdot M \cdot N_T - \sum_{i=1}^{47}\phi_i \cdot \sigma_{He}^i$$
$$\cdot N_{He} \cdot m \cdot a - L \cdot N_{He} \qquad (3)$$

These are reduced to

$$\frac{dN_T(t)}{dt} = \frac{F \cdot N_F + \sum_{i=1}^{47}\phi_i \cdot \sigma_D^i \cdot N_D \cdot m \cdot a}{M}$$
$$- (\lambda_T + \frac{L}{M}) N_T(t)$$
$$+ \frac{\sum_{i=1}^{47}\phi_i \cdot \sigma_{He}^i \cdot m \cdot a}{M} N_{He}(t)$$
$$\frac{dN_{He}(t)}{dt} = \lambda_T \cdot N_T(t)$$
$$- \frac{\sum_{i=1}^{47}\phi_i \cdot \sigma_{He}^i \cdot m \cdot a + L}{M} N_{He}(t) \qquad (4)$$

Table 2. Nomenclature and Parameters.

	Parameters	System	
		Coolant	Moderator
N_F	tritium concentration in replacement heavy water(kg^{-1})	0	0
λ_T	decay constant (sec^{-1})	1.78×10^{-9}	1.78×10^{-9}
L	heavy water loss rate (kg/sec)	1.67×10^{-4}	5.57×10^{-5}
M	totoal heavy water mass (kg)	9.57×10^4	2.62×10^5
m	heavy water irradiated(kg)	6.022×10^3	2.38×10^5
F	heavy water replacement rate (kg/sec)	equal to loss rate	equal to loss rate
N_D	deuterium concentration per unit mass of D$_2$O (D-atoms/kg)	5.968×10^{25}	6.013×10^{25}
ϕ	thermal neutron flux (neutrons/cm^2-sec)	1.235×10^{14}	2.3×10^{14}
σ_D	deuterium capture cross section (10^{-24}cm^2)	3.27×10^{-4}	4.19×10^{-4}
σ_{He}	^{3}He capture cross section (10^{-24}cm^2)	3205.67	4105.70
a	plant capacity factor	0.80	0.80

The nomenclature and the parameters used in the calculations are tabulated in Table 2. The above equations were solved in two ways - one group Runge-Kutta method and 47 group ANISN calculation with BUGLE-93 (Coupled 47 Neutron, 20 Gamma Ray Group Cross Section Library)(Parcons, 1988 ; Oak Ridg National Laboratory, 1994).

4. RESULTS AND DISCUSSIONS

Fig. 1 presents the various tritium concentrations in the coolant, either calculated or measured. It clearly shows that the calculated data including ^{3}He contribution exceed that excluding ^{3}He contribution about 50%. When compared with the measured data, the tritium concentration including the ^{3}He contribution is slightly greater than the measured value at the beginning of plant operation, but agrees well as the operating time increases. The difference between the two tritium concentrations seems to be caused by the frequent overhaul at the early stage of operation, which the current calculation model cannot consider. Fig. 1 also shows that there is no large difference in the results calculated by one-group and 47-groups.
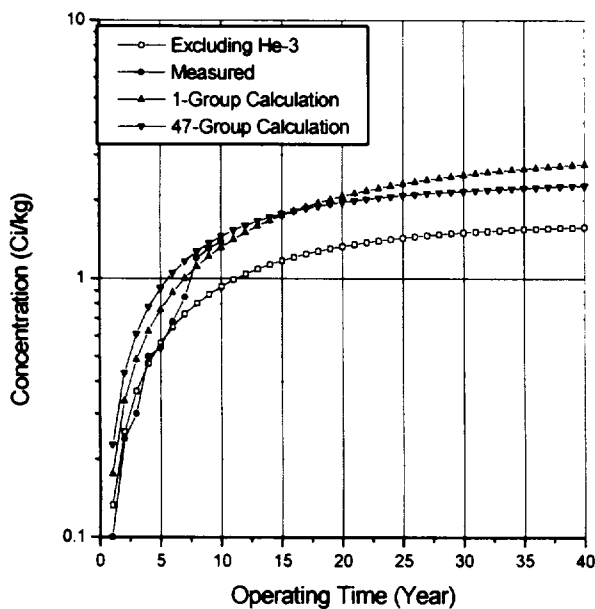

Fig. 1. Tritium concentrations in coolant.

The ratio of the tritium concentration in coolant calculated including ^{3}He contribution to that excluding the contribution, is plotted in Fig. 2. The two curves correspond to 1-group and 47-group calculation, respectively. As indicated, in general the effect of including the ^{3}He contribution is about 50% increase in the amount of tritium accumulation. 1-group calculation underestimates the tritium generation at the early stage of operation and overestimates it at the later stage of operation. While, the trends are reversed in the 47-group calculation.
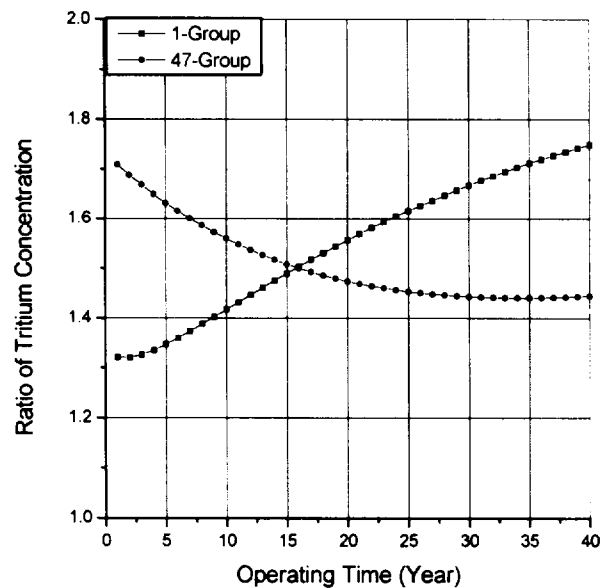

Fig. 2. The ratio of tritium concentration in coolant calculated including the contribution of ^{3}He to that excluding the contribution of ^{3}He.

Similarly, the tritium concentration in the moderator can be also estimated. Calculated tritium concentration and the measured value in moderator are plotted in Fig. 3 as a function of the plant operating time. The tritium concentration in moderator is much larger than that in coolant due to more heavy water under neutron flux and increases from nearly zero at the beginning to an equilibrium value after about 40 years of operation. As shown in Fig. 3, the calculated concentration in moderator is slightly greater than the measured value over the past 10 years of operation.

This discrepancy may result from the idealized model for operating history. Furthermore as we expected earlier, the results also show that ^{3}He contribution to tritium generation in the moderator is negligible.
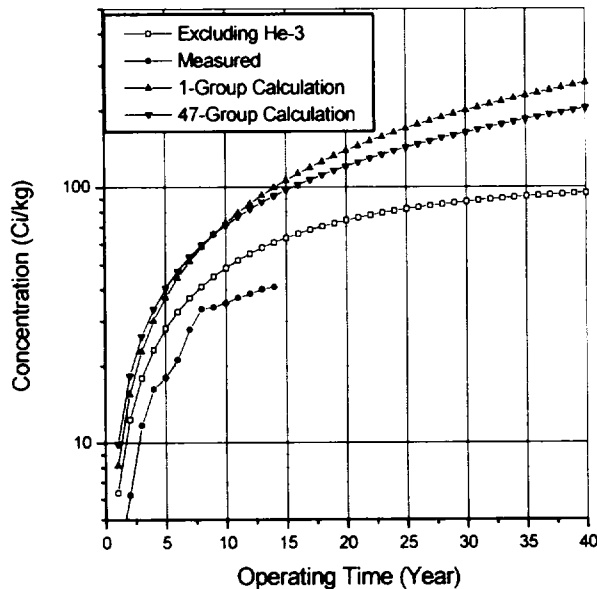

Fig. 3. Tritium concentrations in moderator.

CONCLUSIONS AND RECOMMENDATIONS

The existing calculation models of tritium concentration in PHWR neglect the contribution of ^{3}He both in the moderator and in the coolant due to its relatively low solubility. Based upon the results of this study, we can conclude that the ^{3}He contribution to tritium generation in the moderator is negligible due to its low solubility and the presence of ^{4}He cover gas, but the contribution of ^{3}He to tritium generation in the coolant should be counted. Furthermore we should recognize the ^{3}He contribution gains importance in proportion to the tritium concentration. That means that as the plant ages, the importance of the ^{3}He contribution increases.

In this study, the tritium concentration was estimated with the assumption that the solubility of ^{4}He in light water was regarded as the same as that

of ^{3}He in heavy water. For a more accurate estimation, the use of the solubility of ^{3}He in heavy water is recommended.

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REFERENCES

Sinclair Warren K. 1987. Tritium in the Environment, NCRP Report No.62.

Johnson J. R, R. M. Brown, D. K. Myers, 1992, An Overview of Research at CRNL on the Environmental Aspects, Toxicity, Metabolism and Dosimetry of Tritium, Radiat.Res.50, 197:211.

Harold T. Peterson, Jr and David A. Baker, 1985, Tritium Production, Releases and Populations Doses at Nuclear Power Reactors, Fusion Technology 8, 2544:2550

KEPRI, 1989, Tritium Reduction Research for Wolsong NPP, KEPCO.

Markham Edwin C and Sherman E. Smith, 1995, General Chemistry, Houghton Mifflin Company.

KEPCO, 1976, Wolsong #1 Preliminary Safety Assessment Report Vol.1.

IUPAC, 1979, Solubility Data Series Volume 1:Helium and Neon-gas Solubilities, pp.1~3, Pergamon Press, New York.

IUPAC, 1979, Solubility Data Series Volume 1:Helium and Neon-gas Solubilities, pp.257~260, Pergamon Press, New York.

Kent Parcons D., 1988, ANISN/PC Manual, EG & G Idaho, Inc.

Oak Ridge National Laboratory, 1994, RSIC DATA LIBRARY COLLECTION: Bugle-93, Oak Ridge, Tennessee. 7