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■ 論 文 ■

An Assessment on the Contribution of ^{3}He to the Tritium Generation in the CANDU PHWR

Sung Woo Kwak and Bum Jin Chung*
Research & Development Center, Korea Nuclear Fuel Co., Ltd.
**Nuclear Development Division, Ministry of Science and Technology*

가압중수로에서 헬륨-3이 삼중수소의 생성에 미치는 영향평가

곽성우 · 정범진*
한국원전연료주식회사 · *과학기술처

Abstract—PHWR achieves high neutron economy by adopting heavy water as its moderator and coolant. On the other hand it permits much tritium generation, compared to LWR, due to the neutron capture reaction of deuterium in heavy water. Meanwhile in the reactor core, ^{3}He formed as the result of-decay of tritium, captures a thermal neutron and transforms to tritium again. The existing calculation models on tritium generation in PHWR neglect the contribution of ^{3}He in both moderator and coolant due to its relatively low solubility. However the neutron capture cross-section of ^{3}He is almost 1.6×10^7 times as large as that of deuterium. That means that the dissolved amount of 0.03 ppm of ^{3}He in heavy water is enough to generate the same amount of tritium as that generated by the deuterium of total heavy water in the system. This study dealt with the contribution of ^{3}He to tritium generation. As a sample case, the contribution of ^{3}He to the tritium generation in Wolsong #1 was evaluated and compared to the measured values. According to the result of this study, it is concluded that ^{3}He in coolant contributes very much to the tritium generation but that in moderator shows negligible effects due to the low solubility and ^{4}He cover gas. At the beginning of the plant operation, the contribution of ^{3}He is slightly greater than the measured value but agrees well with the measured as the operating time increases.

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

요약—가압중수로는 감속재와 냉각재로 중수를 채택함으로써 높은 중성자 경제성을 달성하는 대신 중수소의 중성자 포획반응 때문에, 경수로에 비해, 다량의 삼중수소가 발생한다. 한편 원자로심에서, 삼중수소의 β-붕괴결과 발생된 ^{3}He는, 열중성자를 포획하여 다시 삼중수소로 변환된다. 중수로에서 삼중수소의 생성에 대한 기존의 계산모형은, ^{3}He가 상대적으로 낮은 용해도를 가지므로, 그 기여도를 무시해왔다. 그러나 ^{3}He의 중성자 포획단면적은 중수소의 그것에 비해 1.6×10^7 배가 된다. 즉 ^{3}He가 중수내에 0.03

ppm만 녹아있다 하더라도 ^{3}He에 의해 생성되는 삼중수소의 양은 전체 중수에 의한 삼중수소의 양에 필적하게 된다. 본 연구에서는 월성1호기를 대상으로, 중수로에서 ^{3}He가 삼중수소의 생성에 미치는 영향을 평가하였으며 결과를 실측치와 비교하였다. 연구의 결과, 감속재에서는 ^{3}He의 용해도가 낮고 ^{4}He Cover gas 때문에 ^{3}He의 기여도는 무시할 수 있음이 밝혀졌다. 반면 냉각재의 경우 ^{3}He는 삼중수소의 생성에 지대한 영향을 미치는 것으로 나타났다. 또한 본 연구의 계산방법은 원전 운전초기의 냉각재내 삼중수소 생성량은 과대평가 하는 것으로 나타났으나 운전기간이 증가함에 따라 실측치와 잘 일치하는 것으로 나타났다.

중심어 : 삼중수소생성, 헬륨, 용해도, 캔두, 가압중수로

INTRODUCTION

Tritium(^{3}H, denoted as T in this paper), the only radioactive isotope of hydrogen, decays to ^{3}He by 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[1].

Tritium is generated by the reactions of atmospheric atoms with cosmic rays nuclear fission, and activation reactions such as $^{2}H(n, \gamma)T$, $^{3}He(n, p)T$, $^{6}Li(n, \alpha)T$, $^{7}Li(n, n\alpha)T$, $^{9}Be(n, \alpha)^{6}Li$, $^{10}Be(n, \alpha)^{7}Li$, $^{10}Be(n, 2\alpha)T$, $^{11}H(n, \gamma)T$. 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[2, 3]. 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 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[4]. However the neutron capture cross-section of ^{3}He is almost 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).

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[5]. 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[6]

Operating Conditions	Moderator	Coolant
Quantity	264 ton	240 ton
Temperature	70 ℃	310 ℃
Pressure	101.325 Pa	11.1 MPa
Flow rate	940 1/s	94520 1/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, which is 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[7].

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

where X_l : the mole fraction solubility of helium at 101.325Pa(1 atm)

T : temperature

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[6]. 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℃) and 11.1MPa [6]. 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.

$$\frac{n_g}{n_g + n_l} = 1.3956 \times 10^{-3} \text{ and } \frac{W_g}{W_l} = 0.3106 \frac{\text{g-}^4\text{He}}{\text{kg-}H_2O} \quad (4)$$

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[6]. 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.

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

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

$$\frac{d}{dt}(M \cdot N_{He}) = \lambda_T \cdot M \cdot N_T - \sum_{i=1}^{47} \phi_i \cdot \sigma_{He}^i \cdot N_{He} \cdot m \cdot a - L \cdot N_{He}$$

These are reduced to

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

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

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)[9, 10]. Fig. 1 shows the tritium concentration in the coolant.

Table 2 Nomenclature and Parameters used in the calculation.

	Parameters	System	
		Coolant	Moderator
N_F	tritium concentration in replacement heavy water(kg)	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_2O(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	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

As shown in Fig. 1, at the beginning of plant operation, the tritium concentration including the ^{3}He contribution is slightly greater than the measured value but agrees well with the measured value as the operating time increases. The difference between the two tritium concentrations may be caused by the frequent overhaul at the early stage of operation. Fig. 1 also shows that there is no large difference in the results calculated by one-group and 47-groups.

In order to show the ^{3}He contribution explicitly, the ratio of the tritium concentration in coolant calculated including the contribution of ^{3}He to that excluding the contribution, is plotted in Fig. 2.

As indicated in Fig. 2, the ratio begins at about 3 and decreases to about 1.7 after 12 year operation.

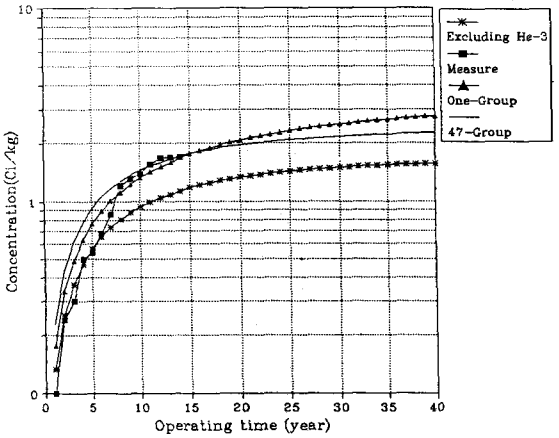

Fig. 1. Tritium concentrations in coolant. (The measured values were obtained by author's personal contact with the KEPCO).

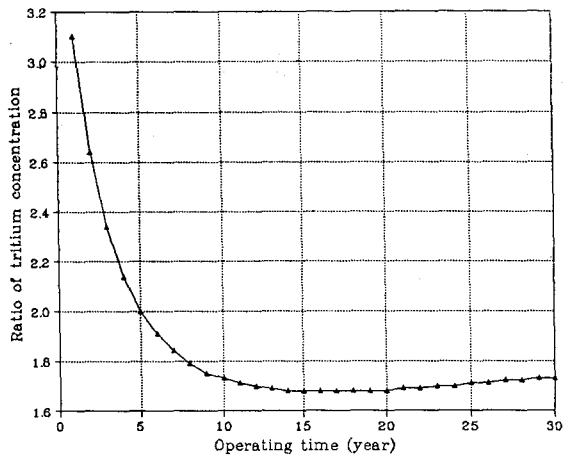

Fig. 2. The ratio of tritium concentration in coolant calculated including the contribution of ^{3}He to that excluding the contribution of ^{3}He.

This can be explained by the coupling of $N_T(t)$ and $N_{He}(t)$ in the equation (4) : it takes about 12 years for tritium and helium-3 concentrations, which began at zeroes, to reach the asymptotic values. But as a whole, the difference between 'one-group' curve and 'excluding He-3' curve increases with plant age and after 40 year operation, it is more than 60%. This is due to the fact that the higher

the tritium concentration is, the higher the ^{3}He concentration is. As a result, ^{3}He makes a larger contribution to the tritium production as the plant ages. Hence it can be concluded that in coolant, the tritium generated by ^{3}He is almost 0.7 times that generated by heavy water. Similarly using the equation (4), 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.

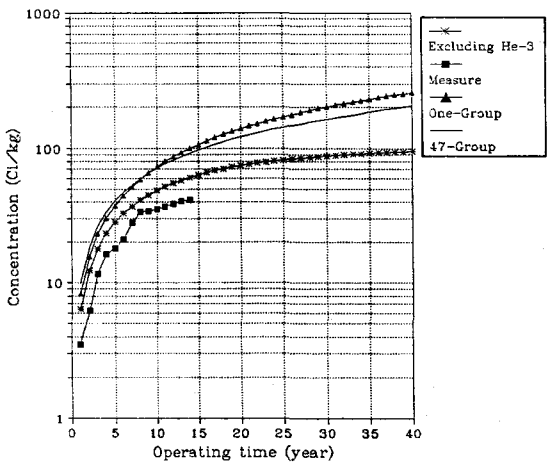

Fig. 3. Tritium concentrations in moderator. (The measured values were obtained by author's personal contact with the KEPCO)

CONCLUSIONS and RECOMMENDATIONS

The existing calculation model of tritium concentration in PHWR neglects 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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