Verification on Tritium Distribution in Hydrogen Production System using High Temperature Gas-Cooled Reactor

Sung Nam Lee*, Nam-il Tak
Korea Atomic Energy Research Institute, DAEDEOK-DAERO 989-111, YUSEONG-GU, DAEJEON, KOREA
Corresponding author: snlee@kaeri.re.kr

1. Introduction

High Temperature Gas-cooled Reactor (HTGR) has been selected as one of the Generation-IV reactors due to its inherent safety. The HTGR is designed to remove a residual heat without an electric supply. Various thermal-hydraulic and system analysis on the HTGR system have been done to investigate a safe and optimized design up to date. However, the research on fission product (FP) distribution in the system may not be enough to meet the practical HTGR designs. There are several types of FPs in the reactor core of HTGR. A tritium among FPs is usually important to predict the amount during the normal operation. Because the coolant of the HTGR is gas phase, the released tritium into the coolant might be easy to leak or permeate to the other system or reactor building. As a result, the tritium transport analysis has been conducted in some countries. Japan Atomic Energy Agency (JAEA, Japan) has developed THYTAN[1] code to analyze the tritium transport in system. Idaho National Laboratory (INL, USA) has developed the Tritium Permeation Analyses Code (TPAC)[2] using the MATLAB S/W. Korea Atomic Energy Research Institute (KAERI, Korea) has been developing Tritium Overall Phenomena analysis(TROPY) code to predict the tritium production and transport in system[3]. Previous studies[4] with TROPY focused on the verification of tritium distribution in the primary loop and the secondary loop of HTGR core. On the present study, the tritium distributions in the loop and the tritium contamination in the product hydrogen were verified with the data by the THYTAN and TPAC code calculations.

2. Methods and Results

The tritium is generated by the several mechanisms. One of main sources is the ternary fission inside the TRISO fuel particle. The others come from the impurities in the core components. The lithium impurities in the graphite block and boron in control rod react with the neutron. The he-3 impurities in the helium coolant also react with the neutron. The generated tritium is remained in the graphite block or released into the coolant by the bound, recoil and diffusion mechanisms. The released tritium into the coolant is easy to leak or permeate into the reactor building or secondary system due to low molecular weight. The HTGR could be connected to the hydrogen product system in order to produce hydrogen. If the tritium permeates to the hydrogen product system through the heat exchangers, the amount of tritium should be assessed to meet regulation limit.

Table I shows the regulation limit by each regulatory body. However, 10 CFR 20 and Korea 2014-34 are based on the maximum release condition to the ambient. The tritium amount by product hydrogen is not defined yet. German and Japan made each limit standard. However, the gap between the values is so large. Therefore, the further consensus is necessary to define the standard.

Table I: Regulation limit

10 CFR 20 Appendix B	*3.7E+3 in Air [Bq/m³]*	*37E+6 in Water [Bq/m³]*
German	*Product Contamination*	*10pCi/g-product*
Japan	*Product Contamination*	*1513 pCi/g-H2*
IAEA	*Exemption Limits*	*27E+6 pCi/g*
EPA	*Drinking water*	*20 pCi/G*
Korea 2014-34, Appendix 5	*HTO*	
	3.E+3 [Bq/m³] in Air	*4.E+7 [Bq/m³] in Water*
	HT	
	2.E+3 [Bq/m³] in Air	*2.E+7 [Bq/m³] in Water*

2.1 Modeling

The tritium distribution in the system of Fig. 1 is obtained by the following one dimensional equation.

$$\frac{\partial C_i}{\partial t} = \dot{q}_{c,i} + \sum_{j=1}^{N_T} a_{i,j}^{*} C_j - \frac{1}{A_F}\frac{\partial}{\partial x}(A_F v C_i) \quad (1)$$

C_i is atoms/m^3, $\dot{q}_{c,i}$ is tritium source by production, release, permeation and so on. v is flow velocity and A_F is the flow area. $a_{i,j}^{*}$ means loss by decay, leak, purification and permeation. The production term is defined as

$$q_{ci} = \sum \alpha_F \frac{dN_T}{dt} + \sum \alpha_{Li6} \frac{dN_{\frac{T}{Li6}}}{dt} + \sum \alpha_{Li7} \frac{dN_{\frac{T}{Li7}}}{dt} + \sum \alpha_{B10} \frac{dN_{T/B10}}{dt} + \sum \alpha_{Be9} \frac{dN_{T/Be9}}{dt}$$

The permeation mechanisms are divided two options, shell and tube and PCHE.

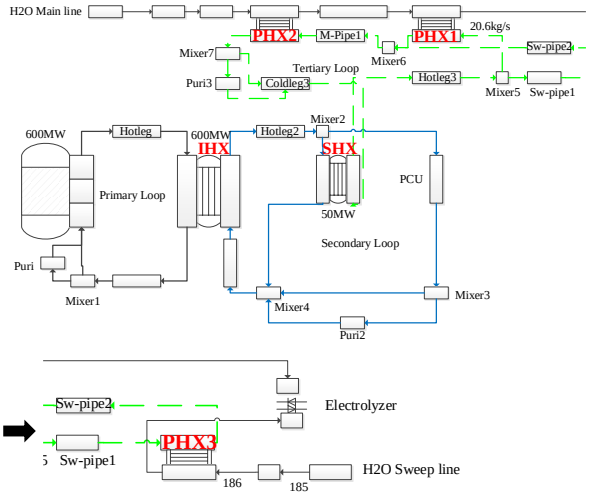

Fig. 1. Schematic of HTGR-HTSE system

2.2 Verification

The components in the HTGR- high temperatures steam electrolysis (HTSE) system were drawn in Fig. 1. The generated heat in the reactor core is transferred to secondary loop by the intermediate heat exchanger (IHX). Some of the heat in the secondary loop is used to generate electricity. The other is transferred to the tertiary loop by the secondary heat exchanger (SHX). The heat in the tertiary loop is again delivered to the hydrogen production system by the process heat exchangers (PHXs). The tritium permeated by the heat exchangers reaches in the hydrogen production system. The transferred tritium will be delivered together with the product hydrogen.

To verify the tritium amount in the HTGR-HTSE hydrogen production system, the TROPY calculation results are compared with the calculation data by the TPAC and THYTAN codes. Each boundary condition for calculation is written in Table II. The designed hydrogen production rate is 7.5E4 m^3(STP)/h.

Table II: HTGR-HTSE boundary conditions

		TPAC	THYTAN
Power [MWt]		600	600
Primary Coolant [°C]		900/658	900/495
Tritium Source [Bq/y]		8.28E13	1.86E14
I H X	Area [m²]	1.35E4	5.039E4
	Thickness [m]	0.02	0.0096
	Pre-exponential Factor [m3(STP)/m/s/$Pa^{0.5}$]	1.36E-10	1.36E-8
	Activation Energy [J/mol K]	5.35E4	7.4E4
S H X	Area [m²]	1.519E3	1.924E3
	Thickness [m]	0.028	0.0282
	Pre-exponential Factor [m3(STP)/m/s/$Pa^{0.5}$]	1.36E-10	1.36E-8
	Activation Energy [J/mol K]	5.35E4	7.4E4
P H X	Area [m²]	34/1287/2161	34/1287/2161
	Thickness [m]	0.01	0.009
	Pre-exponential Factor [m3(STP)/m/s/$Pa^{0.5}$]	1.36E-10	1.36E-10
	Activation Energy [J/mol K]	5.35E4	5.35E4

Table III represents the calculation results by the codes for the HTGR-HTSE system. Overall results agree well. There are discrepancies in the tritium over product hydrogen. The all data for calculation like pipe area, length and etc. was not shown in the each report. Therefore, it has difficulties to compare exactly.

Table III: Calculation results for HTSE - system

Tritium Concentration	TROPY	TPAC
Primary [Ci/m^3(STP)]	2.31E-4	2.48E-4
Secondary [Ci/m^3(STP)]	1.05E-4	1.3E-4
Tertiary [Ci/m^3(STP)]	2.57E-5	2.67E-5
Product Hydrogen [Bq/g-H2]	3.05E+0	6.33E+0
	TROPY	THYTAN
Primary [Ci/m^3(STP)]	1.36E-4	1.15E-4
Secondary [Ci/m^3(STP)]	1.12E-4	1.13E-4
Tertiary [Ci/m^3(STP)]	7.92E-5	1.3E-4
Product Hydrogen [Bq/g-H2]	1.93E+1	2.99E+1

The tritium contamination in the product hydrogen by the TROPY code in the Sulfur Iodine (SI) process (HTGR-SI hydrogen production system) is also compared with the calculated data by the THYTAN code. The primary, secondary and tertiary systems are the same as the conditions of HTSE system. The heat exchangers between the tertiary loop and the SI system were modified. The heat exchanger specifications are explained in Table IV. The calculated results in Table V are similar. The tritium amount on the product hydrogen in the SI system is much higher than that in the HTSE system. The more energy in SI process is needed to supply heat to the decomposer and vaporizer systems.

Therefore, the more heat transfer area in the HX of SI system is required. It resulted in much more permeation of hydrogen from secondary HX to SI process in HTGR-SI hydrogen production system.

Table IV: Heat exchanger at SI - system

	Area [m²]	Thickness [mm]
Decomposer 2	696	0.9
Decomposer 1	696	0.9
Recuperator	696	0.9
Vaporizer 3	696	0.9
Vaporizer 2	696	0.9

Table V: Calculation results for SI - system

Tritium Concentration	TROPY	THYTAN
Primary [Ci/m^3(STP)]	1.11E-4	1.35E-4
Secondary [Ci/m^3(STP)]	6.61E-5	1.35E-4
Tertiary [Ci/m^3(STP)]	4.76E-5	4.77E-5
Tritium in H2 [Bq/g-H2]	2.47E3	1.36E4
Tritium in O2 [Bq/g-O2]	5.07E2	4.67E2

2.3 Design Parameter Analysis

There was still gap in the results in the SI systems. The precise verification would be done if the detail information is available. This section shows the effect of the heat exchanger specification. The parameter of CASE 1 represents the system used in the THYTAN code in the section 2.2. The tritium transport would be limited if the size of heat exchanger can be reduced while maintaining heat balances. The other method is to protect the permeation by coating the surface of the heat exchanger.

Table VI: Heat exchanger parameter at SI - system

		CASE 1	CASE 2	CASE 3
Pre-exponential Factor [m3(STP)/m/s/Pa$^{0.5}$]	IHX	1.36E-8	1.36E-010	1.36E-010
	SHX	1.36E-8	1.36E-010	1.36E-010
Activation Energy [J/mol K]	IHX	7.4E+04	5.35E+04	5.35E+04
	SHX	7.4E+04	5.35E+04	5.35E+04
Area [m²]	IHX	5.039E4	5.039E4	1.35E+04
	SHX	1.924E3	1.924E3	1.52E+03
Thickness [mm]	IHX	0.96	0.960	20
	SHX	2.820	2.820	28.0

Table VII: Calculation results for design parameter

Tritium Concentration	CASE1	CASE2	CASE3
Primary [Ci/m^3(STP)]	1.11E-4	1.73E-4	3.12E-4
Secondary [Ci/m^3(STP)]	6.61E-5	9.85E-5	3.79E-5
Tertiary [Ci/m^3(STP)]	4.76E-5	1.24E-5	4.06E-7
Tritium in H2 [Bq/g-H2]	2.47E+3	6.39E+2	2.09E+1
Tritium in O2 [Bq/g-O2]	5.07E+2	1.32E+2	4.3100

3. Conclusions

The calculated tritium amount in the loop and contamination in the product hydrogen by the TROPY code were verified with the data by the other code calculations in the HTGR – hydrogen production system. The tritium in SI system is much higher than HTSE system. Therefore, more precise analysis is necessary to reduce the tritium amount in the product hydrogen for the SI system. It is necessary to predict the tritium contamination in the product hydrogen with the realistic design parameters like pipe dimension, flow rate, species mass fractions and so on.

Acknowledgements

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (No. 2017M2A8A1014757).

REFERENCES

[1] H. Ohashi, N. Sakaba, T. Nishihara and et al., “Numerical Study on tritium behavior by using isotope exchange reactions in thermochemical water-splitting iodine-sulfer process”, J. Nucl. Sci. Technol., 44(11), p.1407, 2007

[2] E. S. Kim, C. H. Oh, and M. Patterson, “Development of Tritium Permeation Analysis Code and Tritium Transport in a High Temperature Gas-Cooled Reactor Coupled with Hydrogen Production System”, Proceedings of ICAPP 10, 2010

[3] S. N. Lee and N. Tak, “Design Parameter Assessment on the Tritium Release in the VHTR”, KNS Autumn Meeting, Gyeongju, Korea, Oct. 2016

[4] S. N. Lee and N. Tak, “TROPY Code Verification and Validation on Tritium Inventory in a High Temperature Gas cooled Reactor”, KNS Autumn Meeting, Jeju, Korea, May. 2019