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Eung S. Kim
Chang H. Oh
Mike Patterson

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TRITIUM PERMEATION AND TRANSPORT IN THE GASOLINE PRODUCTION SYSTEM COUPLED WITH HIGH TEMPERATURE GAS-COOLED REACTORS (HTGRS)

Eung S. Kim
Idaho National Laboratory
Idaho Falls, Idaho, U.S.A

Chang H. Oh*
Idaho National Laboratory
Idaho Falls, Idaho, U.S.A

Mike Patterson
Idaho National Laboratory
Idaho Falls, Idaho, U.S.A

Abstract-- *This paper describes scoping analyses on tritium behaviors in the HTGR-integrated gasoline production system, which is based on a methanol-to-gasoline (MTG) plant. In this system, the HTGR transfers heat and electricity to the MTG system. This system was analyzed using the TPAC code, which was recently developed by Idaho National Laboratory.*

The global sensitivity analyses were performed to understand and characterize tritium behaviors in the coupled HTGR/MTG system. This Monte Carlo based random sampling method was used to evaluate maximum 17,408 numbers of samples with different input values. According to the analyses, the average tritium concentration in the product gasoline is about 3.05×10^{-3} Bq/cm3, and 62 % cases are within the tritium effluent limit (= $3.7\text{x}10^{-3}$ Bq/cm3[STP]). About 0.19% of released tritium is finally transported from the core to the gasoline product through permeations. This study also identified that the following four parameters are important concerning tritium behaviors in the HTGR/MTG system: (1) tritium source, (2) wall thickness of process heat exchanger, (3) operating temperature, and (4) tritium permeation coefficient of process heat exchanger. These four parameters contribute about 95 % of the total output uncertainties. This study strongly recommends focusing our future research on these four parameters to improve modeling accuracy and to mitigate tritium permeation into the gasoline product. If the permeation barrier is included in the future study, the tritium concentration will be significantly reduced.

I. INTRODUCTION

The high temperature gas-cooled reactor (HTGR) is a helium-cooled, graphite moderated, thermal neutron spectrum nuclear reactor operated at about 8 MPa and higher than 750 ^{o}C. This reactor is envisioned as one of the most promising future energy technology contributed from high efficiency, inherited passive safety features, and its high temperature applications. In particular, this reactor can produce massive hydrogen by splitting water into oxygen and hydrogen, which is clean energy medium without CO_2 release to the environment. For this reason, plenty of international research programs and activities are currently on-going including Gen-IV international forum.

Tritium permeation is an important potential problem that may occur in the HTGR-integrated industrial application systems. Tritium is a hydrogen isotope and primarily generated in the reactor core by ternary fission or various neutron reactions. This species typically exists as a HT molecule in the HTGR. Since this molecule has very small size only about 0.2 nm, it is easily permeated through high temperature metallic surfaces and radioactively contaminates the end-products such as hydrogen and gasoline.

Tritium is a radioactive isotope of hydrogen with the half life of 12.32 years. The nucleus of a tritium atom consists of a proton and two neutrons. Ordinary hydrogen comprises over 99.9% of all naturally occurring hydrogen. The health hazard of tritium is associated with cell damage caused by the ionizing radiation that results from radioactive decay, with the potential for subsequent cancer induction. Nearly all (up to 99%) inhaled tritium can be taken into the body from the lungs, and circulating blood then distributes it to all tissues. Ingested tritium oxide is almost completely absorbed, moving quickly from the gastrointestinal tract to the bloodstream. Within minutes it is found in varying concentrations in body fluids, organs, and other tissues. Generally, tritium is uniformly distributed through all biological fluids within one to two hours. Tritium is eliminated from the body with a biological half-life of 10 days, the same for water. In the U.S., tritium emissions are regulated by the Nuclear Regulatory Commission (NRC) and Environmental Protection Agency (EPA). The suggested regulatory limits are $3.7\text{x}10^{-3}$ Bq/cm^3 in air and 37 Bq/cm^3 in water [1].

Tritium behavior in high temperature gas-cooled reactors (HTGR) has been well evaluated in several countries in the 1970s (e.g., the Dragon reactor in England [2], the Peach Bottom HTGR in the U.S. [3], and Arbeitsgemeinschaft Versuchsreaktor (AVR) in Germany [4]). Data from the operation of HTGRs and from laboratory experiments revealed the mechanism of tritium production, transport, and release to the environment. In the review of tritium behavior by Gainey [5], tritium releases should be well within current federal guidelines for the nuclear plant. For example, the estimated maximum dose to an average adult for a typical 3,000-MWt HTGR with a cooling tower is 0.38 milligram/year, which is slightly more than one tenth of the maximum annual dose allowed [5]. For this reason, no further laboratory scale work on tritium was required at that time. However, their tritium calculations were only concerned with general tritium release and did not examine questions related to nuclear hydrogen production or process steam.

Currently, next generation nuclear program (NGNP) has plans to use the HTGR as a heat source to produce hydrogen or steam for industry or individual users. Ohashi and Sherman [1] estimated steady-state tritium movement and accumulation in an HTGR coupled with hydrogen plants using high temperature steam electrolysis process and thermo-chemical water splitting sulfur-iodine process using the numerical code THYTAN. They also proposed some useful ideas to reduce the tritium levels in the product hydrogen and the other system components on the basis on their analysis results.

This paper is focused on the analyses on the tritium transport and behavior in the HTGR and integrated gasoline production system. In this system, the HTGR transfers heat and electricity to the MTG system for splitting water into oxygen and hydrogen. This system is designed to be operated at the core outlet temperature of 750 ^{o}C with a steam Rankin cycle for power conversion. The current paper summarizes recent tritium research activities achieved in Idaho National Laboratory and shows some notable results.

II. TRITIUM SOURCES AND PATHWAYS

The primary tritium birth mechanism is ternary fission of fuel (e.g., ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu) because of thermal neutrons. Tritium is also generated from ^{6}Li, ^{7}Li, ^{3}He, and ^{10}B by neutron capture reactions in HTGRs as the following:

- ^{6}Li (n, α) ^{3}H,
- ^{7}Li (n, nα) ^{3}H,
- ^{3}He (n, p) ^{3}H,
- ^{10}B (n, 2α) ^{3}H,
- ^{10}B (n, α) ^{7}Li.

^{6}Li and ^{7}Li are impurities in the core graphite material such as the sleeve, spine, reflector, and fuel matrix. ^{3}He is an impurity in the reactor coolant helium. Because helium coolant leaks from the primary loop to the containment vessel, helium is supplied to the primary coolant as a make-up with an impurity of ^{3}He. ^{10}B exists in control rods, burnable poisons, and reflectors.

Figure 1 shows the general tritium pathways in the HTGR system. Tritium is initially generated in the reactor core and about 20~30 % is released to the primary coolant, permeating several barriers of the core. In the primary coolant, some of the tritium is removed by a purification system and some of them can escape outside the coolant by permeation through the components and piping and by leakage with the primary helium coolant. The remaining tritium in the primary coolant permeates through the heat transfer tubes or surfaces of the IHX and gets mixed in with the secondary coolant. In the secondary loop, some of the tritium is removed by the purification system or escapes outside, just as tritium behaves in the primary loop. The remainder of the tritium in the secondary coolant permeates through heat transfer surfaces and transported to the power conversion system or the industrial plants.

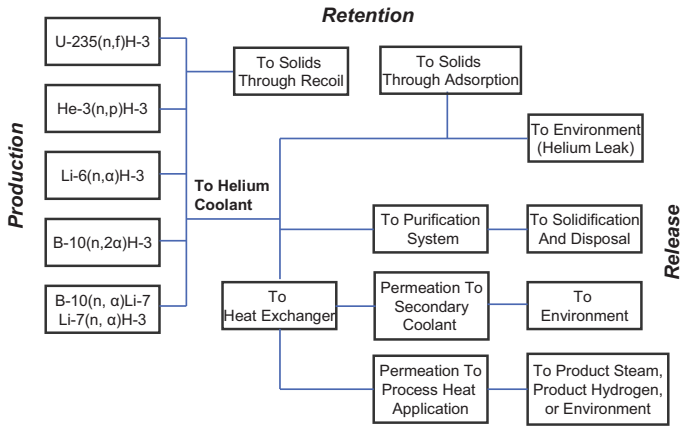

FIGURE 1. GENERAL TRITIUM PATHWAYS IN HTGR.

III. THEORY

This section summarizes the governing equations used for tritium analysis of the HTGR/MTG system. The tritium generation and transport mechanisms in HTGRs are well described by Gainey [5], and Ohashi and Sherman [1].

(a) Mass Balance

The analysis is based on the mass balance equations of tritium containing chemicals and hydrogen. The mass balances of tritium containing chemicals and hydrogen can be expressed as follows:

$$V_j \frac{dC_{i,j}}{dt} = \sum \left(F_{total,j-1} \cdot C_{i,j-1} \right) - F_{total,j} \cdot C_{i,j} - S_{i,j} \quad (1)$$

$$S_{i,j} = -R_{core,i,j} + R_{HX,i,j} + R_{copipe,i,j} + R_{comp,i,j} + R_{leak,i,j} + R_{PF,i,j} + R_{reaction,i,j} \quad (2)$$

(b) Tritium Source

The primary tritium birth mechanism is ternary fission of fuel (e.g., ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu) by the thermal neutrons. The secondary birth mechanisms are from ^{6}Li, ^{7}Li, ^{3}He, and ^{10}B by neutron capture reactions. ^{6}Li and ^{7}Li are impurities in the core graphite material such as the sleeve, spine, reflector, and fuel matrix. ^{3}He is an impurity in the reactor coolant helium. ^{10}B exists in control rods, burnable poisons, and reflectors. Table I summarizes various tritium birth mechanisms and their mathematical representations.

TABLE 1. TRITIUM BIRTH AND RELEASE MODELS.

Tritium Source	Equations
Birth from Ternary Fission	$\frac{d(N_{T(Ter)})}{dt} = K \cdot P \cdot Y - \lambda \cdot N_{T(Ter)}$ (3)
Birth from ^{6}Li (^{6}Li (n, α) ^{3}H)	$\frac{d(N_{Li6})}{dt} = -\phi_{th} \cdot \sigma_{Li6T} \cdot N_{Li6}$ (4) $\frac{d(N_{T(Li6)})}{dt} = \phi_{th} \cdot \sigma_{Li6T} \cdot N_{Li6} - \lambda \cdot N_{T(Li6)}$ (5)
Birth from ^{3}He (^{3}He (n, p) ^{3}H)	$\frac{d(N_{He3})}{dt} = f \cdot N^{\circ}_{He3} - f \cdot N_{He3} - \phi_{He} \cdot \sigma_{He3T} \cdot N_{He3}$ (6) $\frac{d(N_{T(He3)})}{dt} = \phi_{He} \cdot \sigma_{He3T} \cdot N_{He3} - \lambda \cdot N_{T(He3)}$ (7)
Birth from ^{10}B (^{10}B (n, α) ^{7}Li) (^{7}Li (n, nα) ^{3}H)	$\frac{d(N_{B10})}{dt} = -(\phi_{th} \cdot \sigma_{B10Li7} + \phi_f \cdot \sigma_{B10T}) \cdot N_{B10}$ (8) $\frac{d(N_{Li7(B10)})}{dt} = \phi_{th} \cdot \sigma_{B10Li7} \cdot N_{B10} - \phi_f \cdot \sigma_{Li7T} \cdot N_{Li7(B10)}$ (9) $\frac{d(N_{T(B10)})}{dt} = \phi_f \cdot \sigma_{Li7T} \cdot N_{Li7(B10)} + \phi_f \cdot \sigma_{B10T} \cdot N_{B10} - \lambda \cdot N_{T(B10)}$ (10)
Tritium Release Rate	$\frac{N_{T(total)}}{dt} = \sum \left\{ \alpha_{Ter} \frac{d(N_{T(Ter)})}{dt} \right\} + \sum \left[\alpha_{Li} \left\{ \frac{d(N_{T(Li6)})}{dt} + \frac{d(N_{T(Li7)})}{dt} \right\} \right] + \sum \left\{ \alpha_{He3} \frac{d(N_{T(He3)})}{dt} \right\} + \sum \left\{ \alpha_{B10} \frac{d(N_{T(B10)})}{dt} \right\}$ (11) $R_{core,HT,j} = \frac{N_{T(total)}}{dt} \times \frac{1}{N_A} \times \frac{R \cdot T_0}{P_0}$ (12)

(c) Permeation

In the VHTR system, heat exchangers are the main route of tritium permeation. The permeation rate of HT at the heat exchanger, $R_{HX,HT}$ [m^3 (STP)/s] is estimated by the following equation.

$$R_{HX,HT,j}(or \quad R_{copipe,HT,j}) = \frac{A}{l} \cdot k_{p,T} \cdot \left(1 - \frac{P_{H2,l}}{P_{Total,l}} \right) \left(\frac{P_{HT,h}}{\sqrt{P_{H2,h} + P_{HT,h}}} - \frac{P_{HT,l}}{\sqrt{P_{H2,l} + P_{HT,l}}} \right) \quad (13)$$

(d) Leakage

The leak rate of H_2, HT, and HTO with helium leakage in node j, $R_{leak,j}$ is calculated by the following equation:

$$R_{leak,i,j} = V_{total} \cdot L_R \cdot \overline{C}_i \cdot \frac{V_j}{V_{leak}} \quad (14)$$

(e) Purification

The removal rate, R_{PF}, of hydrogen and tritium containing chemicals in the primary and intermediate loop by the purification system is expressed by the following equation:

$$R_{PF,i,j} = F_{PF,He} \cdot \eta_i \cdot C_{i,j} \quad (15)$$

In this study, a complicated electrochemistry model was not considered. Decomposition of H_2O into H_2 and O_2 was calculated by conversion ratios. HTO was assumed to be chemically identical to H_2O.

IV. TRITIUM ANALYSIS ON HTGR/MTG SYSTEM

This section discusses the analyses on the tritium behaviors in the HTGR/MTG system. To analyze the tritium behaviors, this study used TPAC code, which was developed by Idaho National Laboratory [7]. The TPAC is based on the mass balance equations of tritium-containing species and various forms of hydrogen coupled with a variety of tritium sources, sinks, and permeation models shown in the previous section.

The TPAC code was developed using MATLAB and SIMULINK Package by integrating the governing equations. MATLAB is a high-level technical computing program language created by MathWorks. It provides easy matrix manipulation, algorithms implementation, user interface creation, and numerous built-in numerical libraries. SIMULINK is a built-in package integrated in the MATLAB, which is specialized in modeling, simulating, and analyzing multi-domain dynamic systems. The detailed code characteristics and its verifications can be referred to the recent technical report by Oh and Kim [7].

Reference System and TPAC Modeling

This section discusses the tritium analyses results for a coupled HTGR/MTG system (Wood et al. 2010) as shown in Figure 2. This system generates gasoline from natural gas using nuclear heat. In this system, the core outlet temperature is 750°C, and the power conversion system (PCS) is designed based on the steam Rankin cycle. The proposed system includes operation units for air separation (ASU), natural gas purification and reforming (NG-RFMR), methanol synthesis (MEOH-SYN), methanol conversion to gasoline (MTG), power

production (HTSG-ST), the cooling tower, and water treatment. Nuclear heat is used to preheat all streams entering the primary reformer. In addition, nuclear heat is used rather than firing fuel gas for product upgrading in the MTG plant.

Figure 3 shows the TPAC model for the reference HTGR/MTG system. The system configuration, operating temperature, pressure, and flow rates were obtained from McKellar et al. [8]. The component volumes were assumed based on the pre-conceptual design of various VHTR systems.

The IHX sizing and rating were carried out based on the helical-coil tubular heat exchanger and printed circuit heat exchanger (PCHE) designs. The model consists of four different loops; (1) primary helium loop, (2) Secondary helium loop, (3) Power cycle, and (4) methanol-to-gasoline (MTG) system.

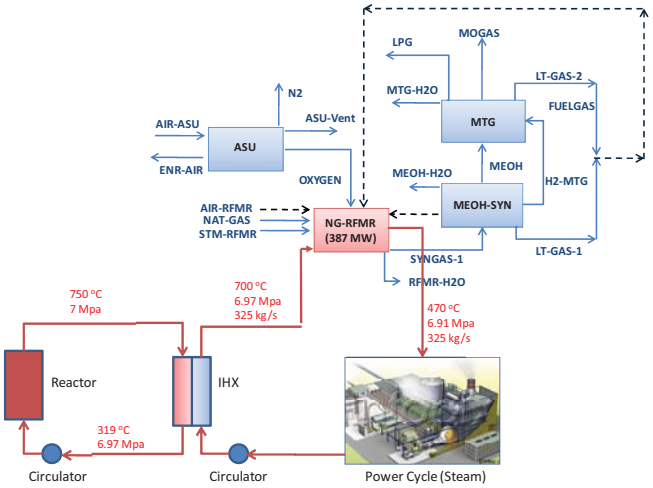

Figure 2. Nuclear-assisted methanol-to-gasoline (MTG) system.

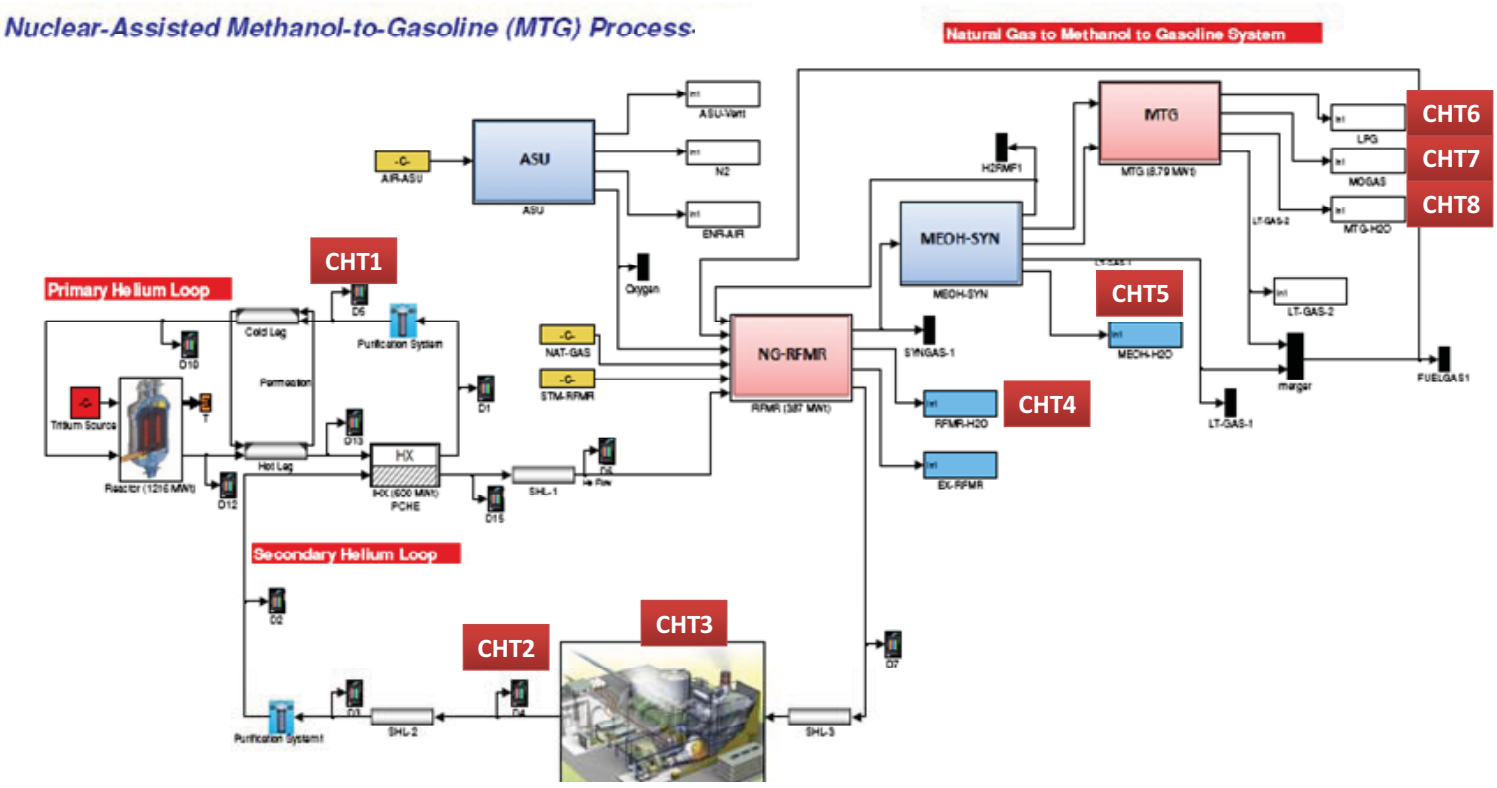

Figure 3. TPAC Model for a coupled HTGR/MTG System.

Analysis Method (Global Sensitivity Analysis)

This study used a global sensitivity analysis method, which is based on the Monte-Carlo based random sampling technique. The global sensitivity analysis is in the category of the sensitivity analysis methods, and it focuses on apportioning the input uncertainty to the uncertainty in the input factors. Typically, global sensitivity analyses are based on the sampling-based method to quantify the influence of uncertain input parameters on the response variability of a numerical model.

There are several different methods that belong to the global sensitivity analysis: standard regression coefficient (SRC), Pearson product moment coefficient (PEAR), Spearman coefficient (SPEAR), measure of importance (i.e. SOBOL, FAST), and etc. [17]. Among them, this study used a variance-based method proposed by Sobol [10], which is based on the variance decomposition. To measure and quantify parameter importance, two sensitivity indices were estimated in this study: first order index and total index.

The first order sensitivity index (S_i), which is called 'main effect', represents the expected amount of variance removed from total output variance, in case that the uncertainty of X_i is known. This measure indicates the relative importance of an individual input variable X_i, in driving the uncertainty, and can be seen as indicating where to direct effort to reduce that uncertainty. The first order indices are very essential parameter for Factor Prioritization (FP) setting, which focuses on indentifying the most important factor.

The total effect for the input variable X_i is the sum of the first order index and all higher order effects in which the factor participates. The total index represents the expected amount of output variance that would remain if X_i is left free to vary over its uncertainty ranges and all other variables having been fixed. The total sensitivity index is the essential parameter for Factors Fixing (FF) setting, which focuses on identifying the factor or the subset of input factors that we can fix at any given values over their ranges of uncertainty without significantly reducing the output variance.

Model Inputs

Tritium behaviors in the HTGR/MTG system are influenced by various factors as described in theory section. This study selected 14 major parameters, which is considered to be important, for uncertainty and sensitivity analyses. Table 2 summarizes the input parameters and their ranges. This study assumed that all the sample distribution is uniform throughout the input ranges, since the detailed information is not available. As described in the previous section, the uniform distribution generally provides more conservative results than the other distributions.

TABLE 2. MAJOR INPUT PARAMETERS AND RANGES.

Parameters	Min	Max	Unit
Tritium Source	1.36E-11	6.44E-11	m^3(STP)/s
Fraction of Mass Flow to Purification System (P-1st)	1.20E-4	2.07E-4	
Fraction of Mass Flow to Purification System (P-2nd)	5.07E-5	8.44E-5	
Reaction Coefficient (IHX)	5.33E-11	1.26E-8	
Reaction Coefficient (S/G)	5.33E-11	1.26E-8	
Reaction Coefficient (Reheater)	5.33E-11	1.26E-8	
Reaction Coefficient (PHX-1)	5.33E-11	1.26E-8	
HX Thickness (IHX)	0.001	0.003	m
HX Thickness (S/G)	0.001	0.003	m
HX Thickness (Reheater)	0.001	0.003	m
HX Thickness (PHX-1)	0.001	0.003	m
HX activation energy (IHX)	5.19E+04	6.4E+04	J/mol
HX activation energy (S/G)	5.19E+04	6.4E+04	J/mol
HX activation energy (Reheater)	5.19E+04	6.4E+04	J/mol
HX activation energy (PHX-1)	5.19E+04	6.4E+04	J/mol
Temperature Level	0.95	1.05	

The tritium source ranges between 1.36×10^{-11} and 6.64×10^{-11} m^3(STP)/s. This condition was obtained from the reported tritium birth rates and the tritium release ratios. The tritium birth rates for the various reactors range from 2.84×10^{-11} Bq/y/MWt (for 3,000 MWt HTGR) to 4.28×10^{-11} Bq/y/MWt (for England 1,500 MWt HTGR). The tritium release ratios are reported to be 0.32 for Peach Bottom reactor and 0.2 for Fort St. Vrain reactor. In the TRIGO code [11], the release ratio is recommended to be 0.1. By combining the tritium birth rates and the release ratios, the tritium release rate (tritium source) was estimated to be ranged between 1.36×10^{-11} and 6.44×10^{-11} m^3(STP)/s. The distribution was assumed to be uniform.

The mass flow into the purification system was determined to be 12 ~ 20 %/h of the total helium inventory in each loop. The numbers, 12 and 20%/h are based on the Peach Bottom reactor and the Fort St. Vrain reactor, respectively.

The tritium permeation coefficients and activation energies for permeation through the HX materials were determined based on the reported Alloy 800 material data. Yang, Baugh, and Baldwin (1977) reported tritium permeation data for the

superheater, evaporater, and economizer of the Peach Bottom reactor. They measured tritium permeation rate after removing surface films formed during reactor operation. Tritium permeation data was also reported by Richards et al. (2006) based on the Peach Bottom steam generator materials. Permeability reported by Richards et al. (2006) includes the effect of surface film formed by steam. Besides the Peach Bottom material test data, lots of laboratory experiments were also conducted using pure Alloy 800 material under well controlled environment (Rohirig et al. (1975), Buckkremer et al. (1978), and Calderoni & Ebner (2010)). This study considered both Peach Bottom experimental data and Lab experimental data for conservative analyses. In the real VHTR operations, oxide films are formed on the surface of the heat exchangers decreasing tritium permeation rates. Generally, oxide film is known to reduce tritium permeation rates, significantly, by 1~3 orders of magnitudes (Sherman and Adams, 2008). Therefore, it should be noted that using the laboratory experimental data (without oxide film) is very conservative approach for analysis of tritium in the VHTR integrated system.

In this analysis, the reference temperature was set to be constant during the analyses. However, the heat exchanger temperatures were determined between 95 and 105 % of the reference temperature. The IHX, SHX, and PHX temperatures were adjusted simultaneously by applying the same multiplication factors, CTL, which represent the temperature level.

In this study, the following six different sampling numbers were taken into considerations for checking convergence and accepting reliability of the results. The minimum number of sample is 544 for Case 1, and the maximum number of sample is 17408 for Case 6.

- Case 1: 544 Samples
- Case 2: 1088 Samples
- Case 3: 2,176 Samples
- Case 4: 4,352 Samples
- Case 5: 8,704 Samples
- Case 6: 17,408 Samples

Results and Discussions

This section summarizes the results of this study focused on the tritium concentrations in the gasoline product of the coupled VHTR/MTG plant. The following three issues are discussed here.

- Tritium concentration in the gasoline product
- Tritium distribution in the HTGR/MTG system
- Important parameters affecting tritium behaviors

(a) Tritium concentration in the gasoline product

Table 3 summarizes the means, standard deviations, and percentiles for the evaluated tritium concentrations in the gasoline products. The average concentration in the product gasoline is 3.05×10^{-3} Bq/cm^3 with 2.46×10^{-3} Bq/cm^3 (~ 54%) in standard deviation.

TABLE 3. MEANS, STANDARD DEVIATIONS, AND PERCENTILES OF TRTIIUM CONCENTRATIONS (Bq/cm^3[STP]) IN THE GASOLINE PRODUCT.

# of Samples	Mean	Std	5% percentile	95% percentile
544	3.25E-03	2.86E-03	6.91E-04	9.12E-03
1088	2.97E-03	2.58E-03	5.22E-04	8.92E-03
2176	3.06E-03	2.50E-03	4.41E-04	8.53E-03
4352	3.02E-03	2.41E-03	4.01E-04	8.08E-03
8704	3.04E-03	2.42E-03	3.51E-04	7.99E-03
17408	3.05E-03	2.46E-03	3.51E-04	8.05E-03

Figures 4 shows the probability distributions of the tritium concentrations in the product gasoline obtained for Case 6 (17,408 samples). According to this figure, the tritium concentration in the product gasoline is widely distributed for the selected input parameters. About 62% of the cases are within the effluent limit (= $3.7\text{x}10^{-3}$ Bq/cm^3[STP]), and 38% exceeds the effluent limit. It indicates that this system has large flexibilities for design conditions and parameters. In addition, the tritium concentration in the gasoline product can be even much more reduced by incorporating the novel tritium permeation barrier methods into the actual heat exchanger design and fabrication.

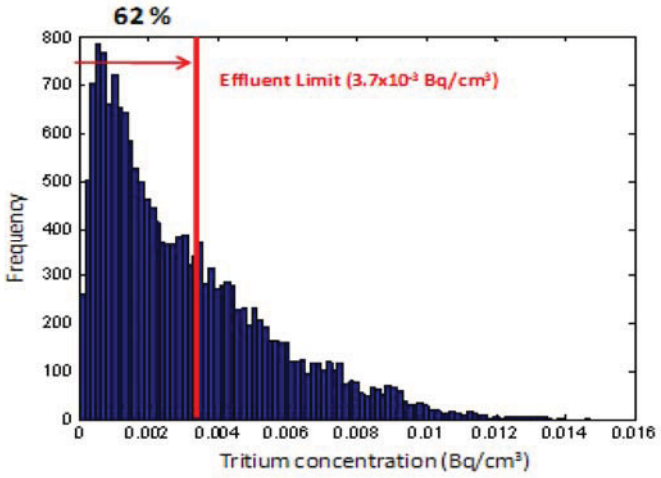

FIGURE 4. TRITIUM CONCENTRATIONS IN THE GASOLINE PRODUCT (CASE 6).

(b) Tritium distribution in the HTGR/MTG system

Figure 5 shows the tritium distributions estimated by the TPAC code in the HTGR/MTG system. According to this result, about 10.4 % of the released tritium from the core is removed by the purification system installed in the primary side. About 10.5 % of the tritium is permeated to the secondary side through the intermediate heat exchanger (IHX) walls. Only 0.27 % of tritium is released to the containment (or

confinement) by helium leakage. In the secondary side, 4.6% of the tritium is removed by the purification system. 4.74 % of tritium is permeated to the steam Rankin cycle. About 0.19 % of tritium transfer to the gasoline production system. According to the calculation results, the purification systems in the primary and secondary system looks the most important for tritium transfer removal in the system. However, the effect of leakage out of the system looks negligible which is less than 0.3% in total. However, the detail numbers can be changed for different system conditions and designs including configuration, temperature, heat exchanger size, and purification system capacity.

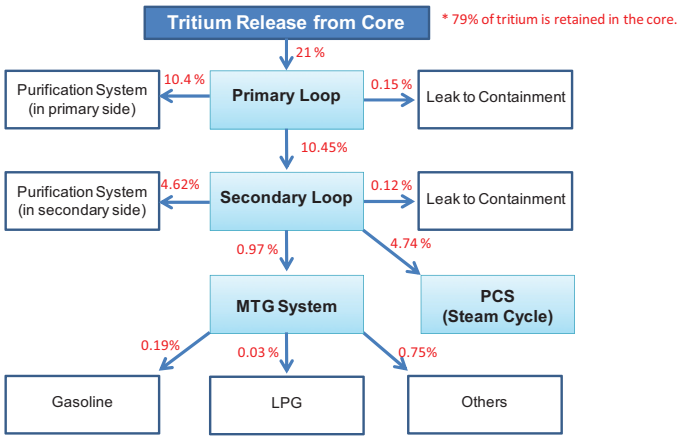

FIGURE 5. TRITIUM DISTRIBUTION IN HTGR/MTG SYSTEM.

(c) Important parameters affecting tritium behaviors in the HTGR/MTG system

This section summarizes the sensitivity analysis results. Two sensitivity indices were used here for measuring parameter importance: (1) first order index and (2) total index.

Main Effect (First Order Sensitivity Index)

The first order index which represents the main effect of an input parameter is closely related to the contribution of the parameter on the total output uncertainties. Table 4 summarizes the results from different numbers of samples. The numbers listed in the table represent the effects of the input parameters on the tritium concentrations in the gasoline product. The main effect of a certain parameter can be quantified by the first-order index. The first-order index is highly related to the modeling uncertainties. If the first-order index of a certain parameter is large, it indicates that that parameter provides large contributions to the output uncertainties. Table 4 summarizes the estimated the first-order indices of the 16 input parameters for tritium concentrations. The locations of the CHT1 through CHT8 are shown in Figure 3. In the sampling-based method, the solution convergence should be checked for different numbers of samples.

TABLE 4. FIRST ORDER SENSITIVITY INDICES.

	CHT1	CHT2	CHT3	CHT4	CHT5	CHT6	CHT7
TS	0.68	0.60	0.24	0.52	0.52	0.52	0.52
CTL	0.03	0.02	0.01	0.02	0.02	0.02	0.02
TH1	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1	0.11	0.09	0.03	0.05	0.05	0.05	0.05
TH4	0.01	0.01	0.05	0.01	0.01	0.01	0.01
PMF1	0.03	0.06	0.16	0.07	0.07	0.07	0.07
C4	0.00	0.00	0.00	0.06	0.06	0.06	0.06
PMF2	0.01	0.00	0.00	-0.01	-0.01	-0.01	-0.01
EA1	0.00	0.00	0.01	0.00	0.00	0.00	0.00
EA4	0.01	0.01	0.03	0.01	0.01	0.01	0.01
TH3	0.00	0.00	0.00	0.00	0.00	0.00	0.00
TH2	0.01	0.01	0.00	0.00	0.00	0.00	0.00
C3	0.01	0.02	0.05	0.01	0.01	0.01	0.01
C2	0.02	0.06	0.14	0.04	0.04	0.04	0.04
EA3	0.00	0.00	0.00	0.01	0.01	0.01	0.01
EA2	0.03	0.02	0.10	0.01	0.01	0.01	0.01

Figure 6 compares the first-order indices for various parameters in the MTG system. As shown in this figure, the TS has the largest first-order index, which is 0.52 (see CHT7 in Table 4). It means that 52% of the output uncertainties are generated by the tritium source model and parameter uncertainties. The second important parameters are the permeation coefficients of PHX, Reheater, and IHX. In this case, the sensitivity indices are 0.07, 0.06, and 0.05, respectively. It means that 20% of the output uncertainties are contributed by the permeation coefficients. On the other hand, effects of the following parameters are negligible: CTL, TH3, EA2, C2, EA4, TH4, EA1, PMF1, TH2, and TH1. This result gives us some ideas to reduce modeling uncertainties, which will eventually provide more design margins. The model uncertainties can be effectively reduced by:

- Improving accuracy of the tritium birth or release models
- Improving accuracy of the input parameters on the tritium birth and release models
- Improving accuracy of the tritium permeation models and parameters

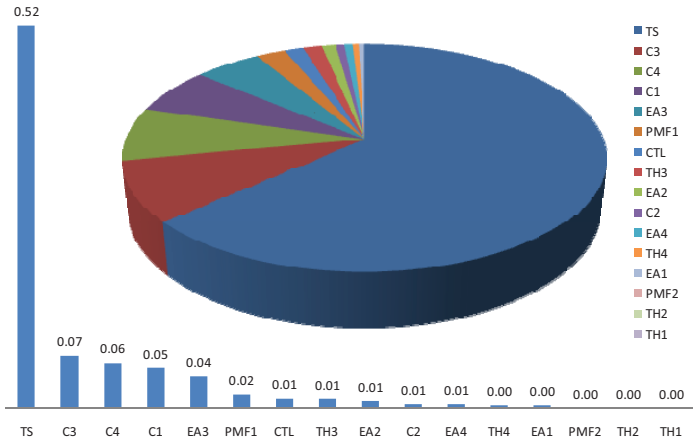

FIGURE 6. FIRST ORDER INDICES.

Total Effect (Total Sensitivity Index)

The total effect of a certain parameter can be quantified by the total index, which is a summation of the first-order index and all its interactions. The total index is highly related to the output value itself. If the total index is large for a certain parameter, it indicates that the output is very sensitive to the change of that parameter. Table 5 summarizes the estimated total indices of the 16 input parameters for tritium concentrations. As shown in this table, the tritium sensitivities in the MTG system are uniform throughout the whole system.

TABLE 5. TOTAL SENSITIVITY INDICES.

	CHT1	CHT2	CHT3	CHT4	CHT5	CHT6	CHT7
TS	0.71	0.64	0.34	0.64	0.64	0.64	0.64
PMF1	0.05	0.03	0.01	0.04	0.04	0.04	0.04
PMF2	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1	0.13	0.12	0.05	0.09	0.09	0.09	0.09
C2	0.01	0.02	0.08	0.02	0.02	0.02	0.02
C3	0.05	0.10	0.24	0.11	0.11	0.11	0.11
C4	0.00	0.01	0.00	0.11	0.11	0.11	0.11
C5	0.01	0.01	0.01	0.01	0.01	0.01	0.01
TH1	0.00	0.00	0.01	0.00	0.00	0.00	0.00
TH2	0.01	0.02	0.04	0.02	0.02	0.02	0.02
TH3	0.00	0.00	0.00	0.01	0.01	0.01	0.01
TH4	0.02	0.02	0.00	0.02	0.02	0.02	0.02
TH5	0.01	0.02	0.08	0.02	0.02	0.02	0.02
EA1	0.04	0.09	0.20	0.09	0.09	0.09	0.09
EA2	0.00	0.00	0.00	0.02	0.02	0.02	0.02
EA3	0.03	0.03	0.14	0.02	0.02	0.02	0.02
EA4	0.71	0.64	0.34	0.64	0.64	0.64	0.64
EA5	0.05	0.03	0.01	0.04	0.04	0.04	0.04
CTL	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Figure 7 compares the total indices for various parameters in the MTG system. As shown in this figure, the TS has the total index, which is 0.64. It means that the change of this parameter affects tritium concentration in the MTG system the most significantly. The second important parameter is the tritium permeation coefficient of the reheater (C4). The third and the fourth important parameters are the permeation coefficient of the PHX and the IHX, respectively. Compared to these four parameters, the effects of the other 14 parameters are very small. This result gives us some ideas to reduce tritium concentration in the MTG system, which will eventually mitigate tritium in the system. The tritium level in the MTG system can be effectively reduced by:

- Improving fuel quality in the core (reducing tritium release)
- Reducing impurities in the core structures (reducing tritium birth)
- Changing or improving heat exchanger wall materials
- Coating heat exchanger walls with tritium barriers

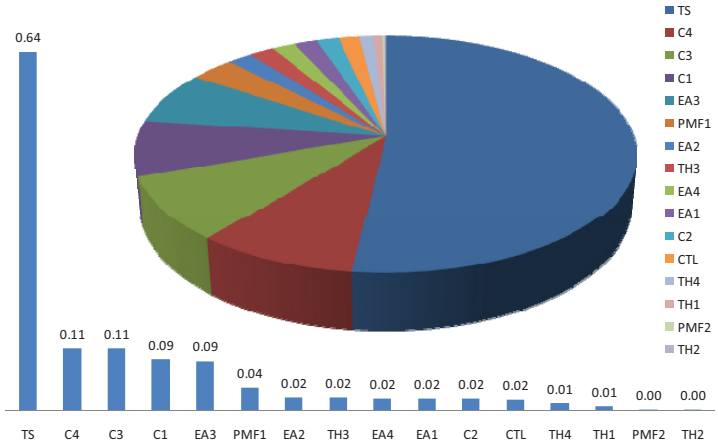

FIGURE 7. TOTAL SENSITIVITY INDICES.

V. SUMMARY AND CONCLUSIONS

In this section, we analyzed tritium behaviors in the improved HTGR/MTG system. The HTGR system was designed based on 721-MWt power and 750°C core outlet temperature. This system consists of the following four separate loops:

- Reactor primary side
- Reactor secondary side
- PCS

- MTG system

The primary heat was transferred to the secondary side through an IHX. In secondary side, the heat was transferred to the PCS and MTG system through an SHX and a PHX. The PCS was based on the steam Rankin cycle. The proposed MTG system includes operation units for air separation (ASU), natural gas purification and reforming (NG-RFMR), methanol synthesis (MEOH-SYN), methanol conversion to gasoline (MTG), power production (HTSG-ST), the cooling tower, and water treatment. Nuclear heat is used to preheat all streams entering the primary reformer.

The HTGR/MTG system was modeled by the TPAC code. Most of the information for modeling was obtained or determined from previous literature, HYSYS flowsheet analyses, and some assumptions. For sensitivity analyses, we agreed on 16 input parameters by discussion and randomly generated 17,408 inputs using the SIMLAB software. For automatic evaluations of a large number of samples, we developed a MATLAB script that links MATLAB workspace parameters and the TPAC input variables. In this study, the following three things were taken into account in detail:

- Tritium concentration in the gasoline product
- Tritium distribution in the HTGR/MTG system
- Important factors affecting tritium behaviors.

First, we estimated tritium concentration in the three locations: (1) gasoline, (2) LPG, and (3) waste water. According to the estimations, the average tritium concentration in the LPG was estimated to be 2.28×10^{-3} Bq/cm^3. In this case, about 75% of the cases were less than the effluent limit (3.7×10^{-3} Bq/cm^3). The average tritium concentration in the gasoline was estimated to be 3.05×10^{-3} Bq/cm^3. About 62 % of the cases were within the tritium effluent limit (3.7×10^{-3} Bq/cm^3).

Second, we estimated tritium distribution in the HTGR/MTG system. According to the analyses, initially, the tritium generated in the reactor core and 20% of the tritium is released to the primary loop. In the primary loop, about 10.4% of the tritium is purified in the purification system, and 10.45% is permeated to the secondary side through the IHX walls. Only 0.15% of the tritium is leaked to the outside of the primary loop. In the secondary loop, 4.62% of the tritium release is purified and 4.74% is permeated to the PCS through the SHX and repeater. About 0.97% of the tritium transports to the MTG system through two PHXs. In the MTG system, the tritium can go everywhere in the system components. Finally, 0.19% of the tritium release goes to the gasoline and 0.03 %, to the LPG. The rest, 0.75%, goes to the other parts of the system components and waste products.

Finally, we conducted global sensitivity analyses in order to identify important factors that affect tritium behaviors in the HTGR/MTG system. These analyses were based on the method proposed by Sobol, and two importance measures were estimated by the SIMLAB software. In these analyses, the following four parameters were identified to be important for both reducing modeling uncertainties and reducing tritium concentration in the gasoline product.

- TS
- Tritium Permeation Coefficient of Reheater (C3)
- Tritium Permeation Coefficient of PHX (C4)
- Tritium permeation coefficient of IHX (C1).

The above parameters were estimated to contribute about 80% of the total uncertainties. It is strongly recommended that future research concentrate on improving those parameters and model accuracies. Also, it is recommended that the methods to effectively control those parameters are developed in the future for mitigating tritium in the system.

ACKNOWLEDGMENTS

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NOMENCLATURE

C_1	Tritium permeation coefficient at IHX
C_2	Tritium permeation coefficient at PHX
C_3	Tritium permeation coefficient at SHX
C_i	average concentration of chemical i in nodes with helium leak
$C_{i,j}$	the volume fraction of chemical *i* in block *j* [m^3 (STP)/m^3 (STP)]
CTL	Fraction of operational temperature to the design condition
EA_1	Activation energy for permeation via IHX
EA_2	Activation energy for permeation via SHX
EA_3	Activation energy for permeation via PHX
f	fractional supply rate of helium coolant [1/s]
$F_{i,eq}$	volumetric flow rate of each chemical at equilibrium [m^3(STP)/s]
$F_{PF,He}$	helium flow rate at purification system [m^3 (STP)/s]
$F_{total,j}$	the volumetric flow rate of all chemicals in block *j* [m^3 (STP)/s]
IHX	Intermediate heat exchanger
IHTL	Intermediate heat transfer loop
HX	Heat exchanger
K	fission rate per thermal megawatt [fission/MW/s]
$k_{p,T}$	permeability of tritium [m^3 (STP)/m/s/Pa$^{0.5}$]
MTG	Methanol-to-gasoline

N_A Avogadro constant

N_{He3} number of ^{3}He atoms

N_{Li6} number of ^{6}Li atoms

N^o_{He3} number of ^{3}He atoms in the supply helium

$N_{T(He3)}$ number of tritium atoms from ^{3}He

$N_{T(Li6)}$ number of tritium atoms from ^{6}Li

$N_{T(release)}$ number of tritium atoms released to the primary coolant

$N_{T(Ter)}$ number of tritium atoms from ternary fission

P reactor power [MW]

PCS Power conversion system

P_0 standard pressure (1.0 1325 × 10^5 Pa).

PHX process side heat exchanger

$P_{HT,h}$ partial pressure of HT at high pressure side [Pa]

$P_{HT,l}$ partial pressure of HT at low pressure side [Pa]

PMF_1 Mass flow fraction to the purification system in the primary side

PMF_2 Mass flow fraction to the purification system in the secondary side

PMF_3 Mass flow fraction to the purification system in the IHTL side

$P_{Total,l}$ total pressure at low pressure side [Pa].

R gas constant (8.314)

$R_{comp,i,j}$ volumetric permeation rate to outside (i = H_2 and HT) [m^3 (STP)/s]

$R_{copipe,i,j}$ volumetric permeation rate at co-axial pipe (i = H_2 and HT) [m^3 (STP)/s]

$R_{core,i,j}$ volumetric release rate from the core to the primary coolant [m^3 (STP)/s]

$R_{HX,i,j}$ volumetric permeation rate at heat exchanger (i = H_2 and HT) [m^3 (STP)/s]

$R_{leak,i,j}$ volumetric leak rate with helium leakage (i = H_2, HT and HTO) [m^3 (STP)/s]

$R_{PF,i,j}$ volumetric removal rate by purification system (i = H_2, HT and HTO) [m^3 (STP)/s]

$R_{reaction,i,j}$ volumetric reaction rate by isotope exchange reactions (i = H_2, HT, HTO, $HTSO_4$ and TI) [m^3 (STP)/s].

SHX Secondary loop heat exchanger

$S_{i,j}$ the total amount of volume change rate of chemical i in block j by considering generation, release, permeation, removal, leakage, and isotope exchange reactions [m^3 (STP)/s]

t time [s]

TH_1 Thickness of the IHX wall

TH_2 Thickness of the SHX wall

TH_3 Thickness of the PHX wall

T_0 standard temperature (273.15 K)

TS Tritium source in Table 4.

V_j volume of block j [m^3 (STP)]

V_{leak} sum of the inventory of nodes with helium leak.

W_{core} helium inventory in core [kg]

W_{total} total primary helium inventory [kg]

Y average yield per fission [1/fission]

α_{B10} fractional release ratio of tritium produced from ^{10}B

α_{He3} fractional release ratio of tritium produced from ^{3}He

α_{Li} fractional release ratio of tritium produced from ^{6}Li and ^{7}Li

α_{Ter} fractional release ratio of tritium produced from ternary fission

η_i fractional efficiency of purification system for removing component i

λ tritium decay constant [1/s].

σ_{He3T} effective cross section for ^{3}He (n, p) T [cm^2]

σ_{Li6T} effective cross section for ^{6}Li (n, α) ^{3}H [cm^2]

Φ_{He} average thermal neutron flux experienced by the total primary helium inventory [n/cm^2/s]

Φ_{th} thermal neutron flux [neutrons/cm^2/s]

REFERENCES

[1] Ohashi, H. and Sherman, S, 2007, Tritium Movement and Accumulation in the NGNP System Interface and Hydrogen Plant, INL/EXT-07-12746, 2007.

[2] Forsyth, N., 1974, "Tritium production and distribution in High Temperature Gas Cooled Reactors: Tritium generation, retention, distribution, and environmental release in a national 1,500 MW(th) HTR," O.E.C.D. High Temperature Reactor Project DRAGON, DP REPORT 905, October 1974.

[3] Wichner, R. P. and F. F. Dyer, 1979, Distribution and Transport of Tritium in the Peach Bottom HTGR, ORNL 5497, Oak Ridge National Laboratory, August 1979.

[4] Steinwarz, W., H. D. Rohrig, and R. Nieder, 1980, Tritium Behavior in an HTR System Based on AVR Experience, IWGGCR2, International Atomic Energy Agency, Vienna (Austria), pp. 153 160.

[5] Gainey, B. W., 1976, A Review of Tritium Behavior in HTGR Systems, GA A13461, General Atomic Company, April 30, 1976.

[6] Atomic Energy Society of Japan, Tritium no Kagaku, Atomic Energy Society of Japan, Vol 31 [in Jananese], 1982.

[7] Oh, C.H and Kim, E.S., Development and Verification of Tritium Analyses Code for Very High Temperature Reactor, INL/EXT-09-16743, 2009.

[8] Mekellar, M.G., Boardman, R.D., Patterson, M.W., Nuclear Assisted Hydrogen Production Analysis, Idaho

National Laboratory, TEV-693, Project Number 23843, 2009.
[9] Satelli, A., Tarntola, S., Campolongo, F., Ratto, M., Sensitivity Analysis in Practice, John Wiley & Sons, ISBN 0-470-87093-1, 2004.
[10] Sobol, L.M., Sensitivity analysis for non linear mathematical models, Math. Model. Comput. Exp., vol 1, pp. 407-414.
[11] General Atomics, 2006, TRITGO Code Description and User's Guide, GA Project 20128, 2006.
[12] Yang, L., W. A. Baugh, and N. L. Baldwin, 1977, Study of Tritium Permeation Through Peach Bottom Steam Generator Tubes, GA A14376, General Atomics Company, June 1977.
[13] Richards, M.B., Shenoy, A.S., Brown, L.C., Buckingham, R.T., Harvego, E.A., Peddicord, K.L., Reza, S.M., and Coupey, J.P., H2-MHR Pre-conceptual Design Report: SI-Based Plant, GA-A25401, General Atomics Company, April 2006.