Development of Tritium Permeation Analysis Code and Tritium Transport in a High Temperature Gas-Cooled Reactor Coupled with Hydrogen Production System

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Abstract – *A tritium permeation analyses code (TPAC) has been developed by Idaho National Laboratory for the purpose of analyzing tritium distributions in Very High Temperature Reactor (VHTR) systems, including integrated hydrogen production systems. A MATLAB SIMULINK software package was used for development of the code. 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. In the TPAC, ternary fission and neutron capture reactions with ^6Li, ^7Li ^{10}B, ^3He were taken into considerations as tritium sources. Purification and leakage models were implemented as main tritium sinks. Permeation of tritium and H_2 through pipes, vessels, and heat exchangers were considered as main tritium transport paths. In addition, electroyzer and isotope exchange models were developed for analyzing hydrogen production systems including high temperature electrolysis and sulfur-iodine (SI) processes.*

Tritium behaviors in a very high temperature reactor/high temperature steam electrolysis (HTSE) system have been analyzed by the TPAC based on the reference indirect parallel configuration investigated by INL in a related VHTR study. This preliminary analysis showed that only 3.5% of tritium released from the core is transferred to the product hydrogen. In this case, the tritium concentration in the hydrogen product is estimated to be 1.04x10^{-3} Bq/cm^3, which is less than the regulatory effluent limit (=3.7x10^{-3} Bq/cm^3) by a factor of 3 to 4. This tritium concentration level can be even reduced further by improved system designs and novel tritium permeation barrier methods.

The TPAC has unlimited flexibility for system configurations and provides an easy method to create models by using a "drag-and-drops" graphical user interface described in this study. Verification of the code was performed by comparisons with analytical solutions and the experimental data from the Peach Bottom Reactor Unit 1 design. Calculations from the THYTAN code developed in Japan and adopted by Japan Atomic Energy Agency were also compared with TPAC results.

I. INTRODUCTION

The Very High Temperature Gas Cooled Reactor (VHTR) is helium-cooled, graphite moderated, thermal neutron spectrum nuclear reactor that will operate at about 8 MPa and 900 ^{o}C. The VHTR is one of six reactor concepts recommended by the Generation IV Technology Roadmap for further development (DOE 2002) and envisioned as one of the most promising future energy technology contributed from high efficiency, inherited passive safety features, and high temperature applications including hydrogen production.

Currently, one potential problem of using VHTRs for high temperature application such as hydrogen production is tritium permeation from the primary coolant to the hydrogen product through heat transfer surfaces. In the VHTRs, the tritium mostly comes from ternary fission of the fuel and neutron capture reactions with lithium impurities in the graphite, boron control materials in the reflector block, and ^{3}He naturally occurring in the helium

coolant. Tritium that enters or is formed in the helium primary coolant will be circulated or permeated to the secondary coolant and the intermediate heat transfer loop. Finally, the permeated tritium enters the high temperature application system and radioactively contaminates the product hydrogen and process steam.

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. This contrasts with the nucleus of an ordinary hydrogen atom and a deuterium atom. Ordinary hydrogen comprises over 99.9% of all naturally occurring hydrogen. Deuterium comprises 0.02% and tritium comprises about a 10^{-16} % of natural hydrogen. The chemical properties of tritium are essentially the same as ordinary hydrogen. Typically, tritium exists as a form of HT (^{1}H-^{3}H) because of isotope exchange reaction between T_2 (^{3}H-^{3}H) and H_2 (Wichner and Dyer 1979). Tritiated water is another common form of tritium. In the tritiated water, a tritium atom replaces one of the hydrogen atoms so the chemical form is HTO rather than H_2O.

Effect of radiation from tritium on health is relatively small and hazard only if it is taken into the body, because tritium decays by emitting a low-energy beta particle with no gamma radiation. This beta particle cannot penetrate deeply into tissue or travel far in air. The most likely form of uptake is as tritiated water. Uptake of tritium gas is typically very low (less than 1%). 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 regulatory limits are 3.7×10^{-3} Bq/cm^3 in air and 37 Bq/m^3 in water (Ohashi and Sherman 2007), respectively.

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 (Forsyth 1972), the Peach Bottom HTGR in the U.S. (Wichner and Dyer 1979), and Arbeitsgemeinschaft Versuchsreaktor (AVR) in Germany (Steinwarz, Rohrig, and Nieder 1980). Data from the operation of HTGRs and from laboratory experiments revealed the mechanism of tritium production, transport, and release to the

environment. In his review of tritium behavior by Gainey's (1976), 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 (Gainey 1976). 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 VHTR as a heat source to produce hydrogen or steam for industry or individual users. For this reason, Ohashi and Sherman (2007) recently estimated steady-state tritium movement and accumulation in an NGNP 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 ideas to reduce the tritium levels in the product hydrogen and the other system components on the basis on their calculations.

This paper focuses on the analyses on the tritium transport and behaviors in the VHTR and integrated hydrogen production system. For this purpose, a dynamic simulation code for tritium analyses in the VHTR system (named as TPAC code) has been newly developed. This paper summarizes the code development and verification activities in Idaho National Laboratory (INL), and also provides some preliminary analyses results for the VHTR/HTSE system.

II. SOURCES AND PATHWAYS OF TRITIUM IN VHTR

II.A. Tritium Source

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 VHTRs as the following:

$$^6\text{Li (n, } \alpha) \, ^3\text{H,} \tag{1}$$
$$^7\text{Li (n, } n\alpha) \, ^3\text{H} \tag{2}$$
$$^3\text{He (n, p) } ^3\text{H} \tag{3}$$
$$^{10}\text{B (n, } 2\alpha) \, ^3\text{H} \tag{4}$$
$$^{10}\text{B (n, } \alpha) \, ^7\text{Li .} \tag{5}$$

^{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.

II.B. Tritium Pathways

Tritium generated in the fuel particles by ternary fissions can escape into a primary coolant, permeating several barriers of the fuel particles. In addition, tritium born from [10]B and [6]Li can pass into the primary coolant. The principal chemical form for tritium in the reactor coolant was reported as HT ([1]H-^{3}H) because of the isotope exchange reaction between T_2 ([3]H-^{3}H) and H_2 (Wichner and Dyer 1979). Some of the tritium in the primary coolant is removed by a purification system installed in the primary loop. Some of the tritium 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 intermediate heat exchanger (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 gets mixed into the tertiary coolant. Transportation of tritium into the tertiary coolant is the same as for the secondary coolant. It permeates through the heat transfer surfaces of the process heat exchangers and is mixed into the process chemicals of the hydrogen plant.

Tritium that has permeated the tertiary loop connected to the hydrogen plant can react with hydrogen-containing process chemicals through isotopic exchange reactions. For example, the high-temperature steam electrolysis (HTSE) process contains H_2O. Therefore, HTO (tritiated water) is produced by the isotope exchange reaction between HT and H_2O. Gaseous HTO and HT escape from the HTSE process with the product hydrogen and oxygen. A part of the liquid HTO flows out from the hydrogen plant with the drain water. The remainder of the liquid HTO circulates into the plant with the recycling water and accumulates in the water of the HTSE process. The SI process used in hydrogen production also contains H_2O, H_2SO_4, and HI chemicals from which HTO, $HTSO_4$, and TI may be produced through isotope exchange reactions (Ohashi and Sherman, 2007). These tritium-containing chemicals circulate with and accumulate in process the chemicals. Only gaseous HT and HTO can escape from the SI process with the product hydrogen and oxygen.

The above tritium pathways in VHTRs are well illustrated in Fig. 1. All of the production mechanisms lead to either retention in solids or entry into coolant systems. The tritium in the primary coolant can be adsorbed on

graphite surface, be leaked out of the primary circuit, be removed in the purification system, or be permeated to the process heat application system like hydrogen production.
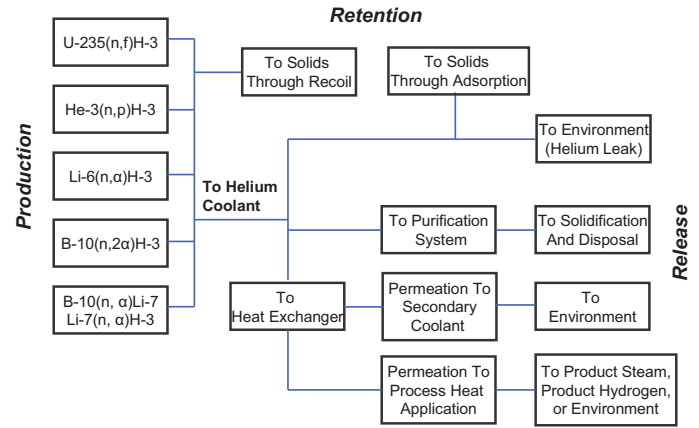

Fig. 1. General Tritium Pathways in VHTR.

III. DEVELOPMENT OF TRITIUM ANALYSES CODE AND VERIFICATION

III.A. Theory

This section summarizes the theory adopted in the TPAC code developed by INL for analyses of tritium behaviors in the VHTR system. The TPAC code is capable of modeling the tritium dynamic behaviors in the simplified representations of the VHTR, the HTSE process, the SI process, and other process application systems.

(a) Mass Balance

The TPAC code 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:

$$V_j \frac{dC_{i,j}}{dt} = \sum \left(F_{total,j-1} \cdot C_{i,j-1} \right) - F_{total,j} \cdot C_{i,j} - S_{i,j} \quad (6)$$

$$S_{i,j} = -R_{core,i,j} + R_{HX,i,j} + R_{copipe,i,j}$$
$$+ R_{comp,i,j} + R_{leak,i,j} + R_{PF,i,j} + R_{reaction,i,j} \quad (7)$$

(b) Tritium Source

The tritium generation mechanism in VHTRs is well described by Gainey (1976), and Ohashi and Sherman (2007). 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 I. Tritium Birth and Release Models.

Tritium Source	Equations	
Birth from Ternary Fission	$\dfrac{d(N_{T(Ter)})}{dt} = K \cdot P \cdot Y - \lambda \cdot N_{T(Ter)}$	(8)
Birth from ^{6}Li (^{6}Li (n, α) ^{3}H)	$\dfrac{d(N_{Li6})}{dt} = -\phi_{th} \cdot \sigma_{Li6T} \cdot N_{Li6}$	(9)
	$\dfrac{d(N_{T(Li6)})}{dt} = \phi_{th} \cdot \sigma_{Li6T} \cdot N_{Li6} - \lambda \cdot N_{T(Li6)}$	(10)
Birth from ^{3}He (^{3}He (n, p) ^{3}H)	$\dfrac{d(N_{He3})}{dt} = f \cdot N^{\circ}_{He3} - f \cdot N_{He3} - \phi_{He} \cdot \sigma_{He3T} \cdot N_{He3}$	(11)
	$\dfrac{d(N_{T(He3)})}{dt} = \phi_{He} \cdot \sigma_{He3T} \cdot N_{He3} - \lambda \cdot N_{T(He3)}$	(12)
Birth from ^{10}B (^{3}He (n, p) ^{3}H) (^{10}B (n, α) ^{7}Li) (^{7}Li (n, nα) ^{3}H)	$\dfrac{d(N_{B10})}{dt} = -(\phi_{th} \cdot \sigma_{B10LiT} + \phi_f \cdot \sigma_{B10T}) \cdot N_{B10}$	(13)
	$\dfrac{d(N_{Li7(B10)})}{dt} = \phi_{th} \cdot \sigma_{B10LiT} \cdot N_{B10} - \phi_f \cdot \sigma_{LiTT} \cdot N_{Li7(B10)}$	(14)
	$\dfrac{d(N_{T(B10)})}{dt} = \phi_f \cdot \sigma_{LiTT} \cdot N_{Li7(B10)} + \phi_f \cdot \sigma_{B10T} \cdot N_{B10} - \lambda \cdot N_{T(B10)}$	(15)
Tritium Release Rate	$\dfrac{N_{T(total)}}{dt} = \sum \left\{ \alpha_{Ter} \dfrac{d(N_{T(Ter)})}{dt} \right\} + \sum \left[\alpha_{Li} \left\{ \dfrac{d(N_{T(Li6)})}{dt} + \dfrac{d(N_{T(Li7)})}{dt} \right\} \right]$ $+ \sum \left\{ \alpha_{He3} \dfrac{d(N_{T(He3)})}{dt} \right\} + \sum \left\{ \alpha_{B10} \dfrac{d(N_{T(B10)})}{dt} \right\}$	(16)
	$R_{core,HT,j} = \dfrac{N_{T(total)}}{dt} \times \dfrac{1}{N_A} \times \dfrac{R \cdot T_0}{P_0}$	(17)

(c) Permeation

In the VHTR system, heat exchangers are the main route of tritium permeation. Therefore, good predictions on the permeation rates through the heat transfer surfaces play very important roles for estimating the tritium distributions and contamination levels in hydrogen product. The permeation rate of HT at the heat exchanger, $R_{HX,HT}$ [m^3 (STP)/s] is estimated by considering the effect of the existence of hydrogen on the adsorption-dissociation and recombination-desorption step as follows:

$$R_{HX,HT,j} \left(\text{or } R_{copipe,HT,j} \right)$$
$$= \frac{A}{l} \cdot k_{p,T} \cdot \left(1 - \frac{P_{H2,l}}{P_{Total,l}} \right) \left(\frac{P_{HT,h}}{\sqrt{P_{H2,h} + P_{HT,h}}} - \frac{P_{HT,l}}{\sqrt{P_{H2,l} + P_{HT,l}}} \right) \quad (18)$$

(d) Leakage

The leak rate of H$_2$, HT, and HTO with helium leakage in node j, $R_{leak,j}$ is calculated by using the following equation:

$$R_{leak,i,j} = V_{total} \cdot L_R \cdot \overline{C}_i \cdot \frac{V_j}{V_{leak}} \quad (19)$$

(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:

$$R_{PF,i,j} = F_{PF,He} \cdot \eta_i \cdot C_{i,j} \quad (20)$$

(f) Electrolyzer

In the TPAC code, a simple electrolyzer model has been implemented in order to match the species conservation for the following chemical reaction.

$$H_2O = H_2 + 0.5O_2 \quad (21)$$

In this model, a complicated electrochemistry model has not been considered. Decomposition of H$_2$O into H$_2$ and O$_2$ was calculated by conversion ratios. HTO was assumed to be chemically identical to H$_2$O.

(g) Isotope Exchange

The SI process contains H$_2$, H$_2$O, H$_2$SO$_4$, and HI and the HTSE process contains H$_2$ and H$_2$O as hydrogen-containing chemicals. The primary and intermediate coolants also contain H$_2$ and H$_2$O as impurities. To analyze the tritium behaviors in the VHTR/Hydrogen production system, we need some additional models that can handle isotope exchange reactions between some chemicals. The following isotope exchange reactions between tritium containing chemicals and hydrogen containing chemicals were implemented into the TPAC code. However, this paper is focused on the HTSE.

$$HT + H_2O = H_2 + HTO \quad (22)$$
$$HT + H_2SO_4 = H_2 + HTSO_4 \quad (23)$$
$$HT + HI = H_2 + TI \quad (24)$$
$$HTO + H_2SO_4 = H_2O + HTSO_4 \quad (25)$$
$$HTO + HI = H_2O + TI \quad (26)$$

The TPAC code used simple equilibrium models for the isotope exchange reactions. The equilibrium constants for above reactions were referred to the reports by Atomic Energy Society of Japan (1982) and by Ohashi and Sherman (2007).

III.B.TPAC Code Development and Structure

The TPAC code was developed by using MATLAB/SIMULINK Package (Mathwork 2009) for integration of 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 multidomain dynamic systems. Its primary interface is a graphical block diagramming tool and a customizable set of block libraries. It offers tight integration with the rest of the MATLAB environment. It supports linear and nonlinear system modeled in continuous time, sampled time, or hybrid of both.

The TPAC code has been developed based on the SIMULINK user interface. All the TPAC component libraries were built in the SIMULINK model library so that the TPAC component blocks can be easily integrated with pre-existing SIMULINK model blocks. It provides enormous flexibility and extensibility to the code. The TPAC code provides large flexibility for system configurations with very convenient graphical user interface (GUI). Any configurations of the systems (not only VHTR but also general components) can be made by easy drag-and-drops on the screen. Fig. 2 shows the basic TPAC workspace and GUI.
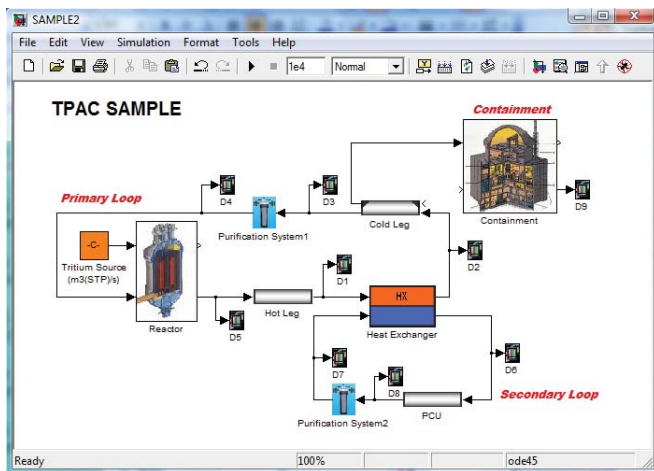

Fig. 2. TPAC code GUI (workspace).

The TPAC code has a variety of library categories. Each category has different sets of component block models. Fig. 3 shows the structure of the TPAC code component libraries. The current version of the TPAC code is 1.5b, and it consists of twelve different categories as shown in the figure. **VHTR System Component** contains the component blocks related to the VHTR systems such as core and vessel. **Pipe Component** contains various types of pipe component blocks including VHTR, HTSE, and SI systems. **Heat Exchanger Component** contains two different heat exchanger component blocks. **Vessel Component** contains various vessel components related to VHTR, HTSE, and SI systems. **Containment Component** contains the reactor containment block. **Purification System Component** contains a tritium purification system block. **Electrolyzer Component** contains an electrolyzer block. **Flow Distributions Component** contains flow distributor and mixer blocks. **Boundary Flow Component** contains a bound flow block which can be used for open-flow or fixed flows. **Detector Component** contains a detector block which can display the flow conditions during or after simulation. **Data Save Component** contains data save blocks which export calculated results to MATLAB Workspace or external files. **Sample Simulation** contains some sample simulation cases including those used for code verification works.
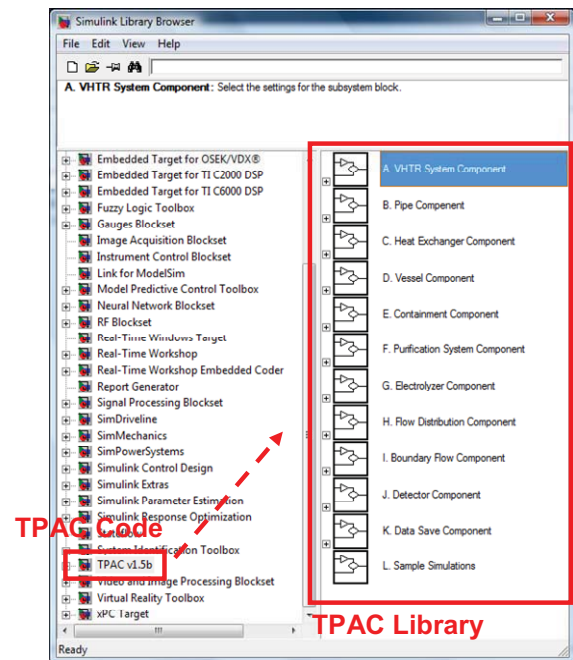

Fig. 3. TPAC code GUI (component library window).

III.C. Verification of the TPAC Code

Verification of the TPAC code has been performed by analytical solutions and reported data based on the Peach Bottom reactor (Wichner and Dyer, 1979). The Peach Bottom reactor was a helium-cooled graphite-moderated reactor built in the United States in 1958. In this reactor, the primary circuit consisted of two loops, each containing a helium compressor and steam generator. The total helium flow of 210,000 kg/h was divided equally between the two loops. Coolant temperatures at the core inlet and outlet of the reactor vessel were 345 and 714°C, respectively, and the primary loop pressure was approximately 2.4 MPa (335

psig). The reactor and steam generator were connected by a concentric duct. Coolant exiting the reactor flows in the inner pipe of the concentric duct. The steam generators were forced recirculation, drum type boilers that have pendant U-tube economizer, evaporator, and superheater sections.

The verification has been performed in two ways. First, the separate models (e.g. tritium birth models, leakage model, permeation model, purification system model, ion-exchange model, and etc.) were verified independently. After all the separate models were successfully verified, the total system model has been verified based on the Peach Bottom Core 2 operation designed to be BISO (fuel coated with two ceramic layers) in order to retain gaseous fission products during the life (i.e. from July 1970 to October 1974). According to the verification results, the TPAC calculation results showed very good agreement with the analytical and experimental data as a whole. This paper summarizes only some of the results among them. For more information, it is strongly recommended to be referred to the technical report published by INL (Oh and Kim, 2009).

Tritium source models have been first verified against analytical solution, reported results, and the THYTAN code results. The THYTAN code is a numerical analysis code for tritium analyses, which was developed by Japan Atomic Energy Research Institute in the late 1990s (Ohashi and Sherman 2007). TABLE II summarizes the comparisons between the TPAC code solutions and other reference results for various tritium birth types.

According to the Table II, The TPAC code result shows very good agreement with the reported values and the analytical solutions for the ternary fission. The reported tritium birth by ternary fission during 1,550 days was 1,210 Ci (= 4.43×10^{13} Bq) and the TPAC solution is 4.46×10^{13} Bq. It is also in good agreement with the THYTAN numerical solution (= 4.42×10^{13} Bq).

For the tritium birth from Li impurities, there were some unknown discrepancies between reported values and analytical solutions that have not yet been revealed. However, the TPAC solutions showed good agreement with the analytical solutions and THYTAN results for various regions of the core. It verifies that the TPAC code correctly solves the governing equations.

For the tritium birth from ^{3}He, the TPAC solutions showed good agreement with the analytical solutions and the THYTAN results for the Core 2 operation in the Peach Bottom reactor. It also verifies the TPAC code for the tritium birth from ^{3}He. However, the reported values by Wichner and Dyer (1979) are a little bit larger than both analytical and computed solutions within 10–60% as

Ohashi and Sherman (2007) already reported. The reason is still unknown yet.

Tritium from ^{10}B is produced in the poisoned spine and in the control rod. However, since the boron level and control rod position varies with time in the control rod, only poisoned spine was taken into consideration here. According to the results, the TPAC solution shows very good agreement with the analytical solutions and the THYTAN results.

TABLE II. Comparisons of tritium activity at the Core 2 operation of the Peach Bottom reactor.

Graphite Component	Activity			
	Reported Value (Wichner and Dyer 1979)	Analytical Solution	THYTAN (Ohashi and Sherman 2007)	TPAC Solution
	(Bq)	(Bq)	(Bq)	(Bq)
Birth from Ternary Fission				
Fuel	4.43×10^{13}	4.43×10^{13}	4.42×10^{13}	4.43×10^{13}
Birth from ^{6}Li, ^{7}Li				
Sleeve	5.13×10^{11}	5.12×10^{11}	5.12×10^{11}	5.12×10^{11}
Spine	3.66×10^{10}	3.78×10^{10}	3.78×10^{10}	3.78×10^{11}
Removal radial reflector	6.01×10^{11}	5.76×10^{11}	5.75×10^{11}	5.76×10^{11}
Permanent radial reflector	6.89×10^{11}	6.72×10^{11}	6.69×10^{11}	6.68×10^{11}
Axial reflector	3.37×10^{11}	3.42×10^{11}	3.42×10^{11}	3.42×10^{11}
Fuel matrix	4.80×10^{11}	5.68×10^{11}	5.68×10^{11}	5.68×10^{11}
Birth from ^{3}He				
In sleeve graphite	1.98×10^{11}	1.31×10^{11}	1.26×10^{11}	1.28×10^{11}
In removal radial reflector	4.95×10^{11}	3.20×10^{11}	3.09×10^{11}	3.12×10^{11}
In permanent radial reflector	5.68×10^{11}	5.43×10^{11}	5.24×10^{11}	5.31×10^{11}
In axial reflector	1.14×10^{12}	9.17×10^{11}	8.86×10^{11}	8.98×10^{11}
Birth from ^{10}B				
Poisoned Spine	3.14×10^{12}	3.19×10^{12}	3.18×10^{12}	3.19×10^{12}

Verification of the TPAC code has also been carried out against Peach Bottom reactor Core 2 operation. Fig. 4 shows the TPAC modeling for the whole system of the Peach Bottom reactor. The Peach Bottom reactor is composed of a reactor, two steam generators, two concentric pipes, containment, and two purification systems (Compare et. al. 1974). However, for simplicity, two steam generator loops has been simplified to be a single steam generator loop in the modeling. Tritium flow path of this system is as follows. First, the tritium is generated in the reactor core and released. Majority of the released tritium enters into the main flow distributed in the whole system. The rest of the tritium from core is purged to the purification system for removing tritium from the
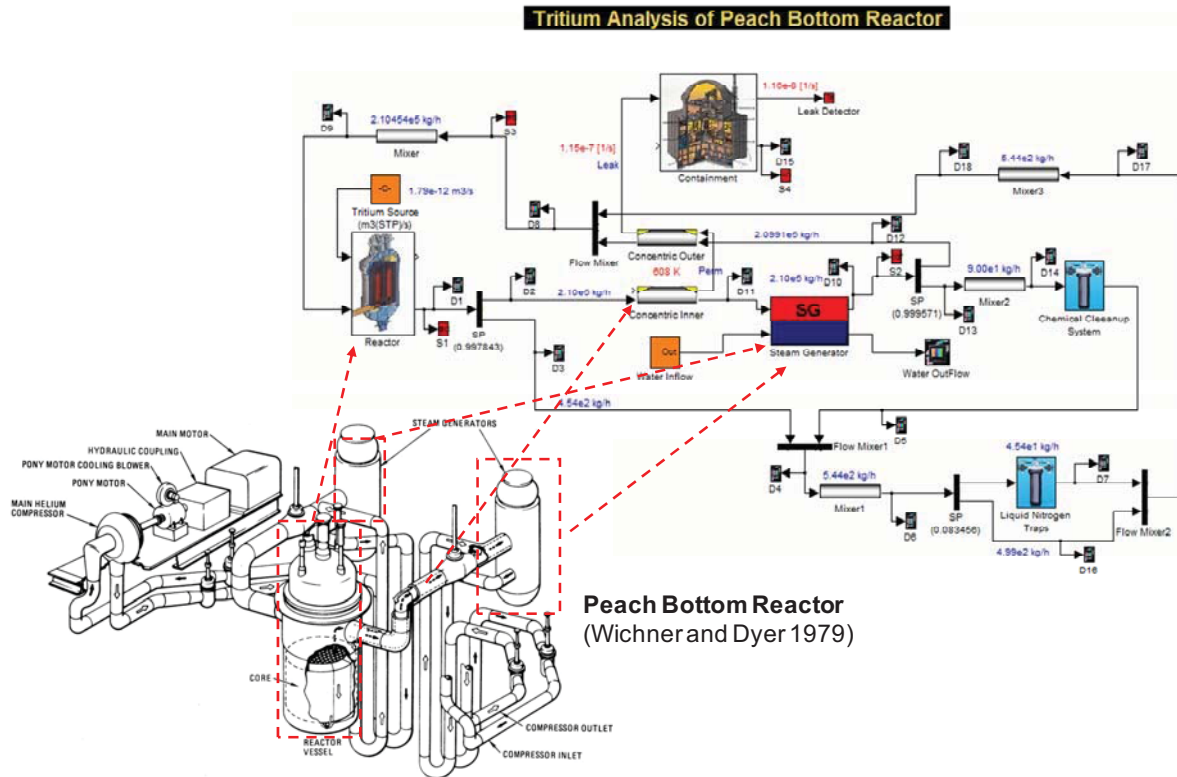
Fig. 4. TPAC modeling for Peach Bottom reactor.

primary loop. Some of the tritium in the main flow is permeated to secondary side through the steam generator walls or leaked to the containment through the pipe lines. A small portion of main flow is purged to another purification system. The total volume of the system was set to be 232 m^3, and the size of the each component was assumed based on the THYTAN code input developed by Ohashi and Sherman (2007). The temperature and the pressure of all the components were set as 809 K and 23 atm, which are the average temperature and pressure in the system.

The tritium birth rate of the core was determined between 310 Ci (1.15×10^{13} Bq) in minimum and 1203 Ci (4.45×10^{13} Bq) in maximum for 1,550 days of Core 2 operation. The tritium birth rate, 310 Ci is the leakage to the fission product trapping system via the fuel element purge flow, and the value, 1203 Ci is the maximum available release rate of tritium from the core. The tritium birth rate was assumed to be constant during the operating time.

The steam generator was modeled to be a single loop by merging two steam generators into one. The steam generator model consisted of three components; super heater, evaporator and economizer. Each component (superheater, evaporator and economizer) was modeled by 5 nodes. The metal temperatures of the steam generator were determined based on the reactor operating conditions.

The parameters for the permeation models were chosen from the literature by Yang, Baugh, and Baldwin (1977).

Two purification systems were considered in the modeling; (1) Nitrogen trap and (2) Chemical cleanup system. The efficiencies for the system were assumed to be 100 % (Wichner and Dyner 1979). The mass flow-rates for the nitrogen trap and chemical clean-up system were set as 45.4 kg/h and 90.7 kg/h, respectively. The leakage was modeled at the outer pipe of the concentric duct.

The TPAC code estimated the tritium concentration in the primary coolant to be 8.88×10^{-5} $\mu Ci/cm^3$ based on the activity of 310 Ci to the purge loop and 0 Ci to the main flow channel (i.e., Case of minimum release rate) for the average temperature and pressures in the primary side. As for the case of the tritium activity, 310 Ci to the purge flow and 893 Ci to the main flow channel (i.e., Case of maximum release rate), tritium concentration in the primary coolant was calculated to be 3.44×10^{-4} $\mu Ci/cm^3$. The above results are in good agreement with the THYTAN results (i.e. 8.16×10^{-5} $\mu Ci/cm^3$ for minimum and 3.38×10^{-4} $\mu Ci/cm^3$ for maximum), which modeled two steam generator loops and two core regions separately. Fig. 5 shows the comparisons between the reported experimental results (Wichner and Dyer 1979) and two calculated solutions by the THYTAN and the TPAC code. According to the experimental results, the tritium activity in the primary coolant (i.e. main loop HT and HTO)

fluctuated from 2×10^{-6} µCi/cm³ to 3×10^{-4} µCi/cm³ during the operation. As shown in this figure, the computed solutions are in the range of the experimental data, but located at the higher ranges of experimental data. It indicates that the computed solution from the codes (THYTAN and TPAC) is slightly conservative; however, the codes could calculate tritium concentrations at the same order as the experimental results of the Peach Bottom reactor. The calculated TPAC solutions showed good agreement with the THYTAN results even though they used different nodalizations. It is contributed from relatively slow permeation process of the tritium compared to the convection process in the system. This agreement also verifies that the integration and mass conservation models in the TPAC code are correctly solved.
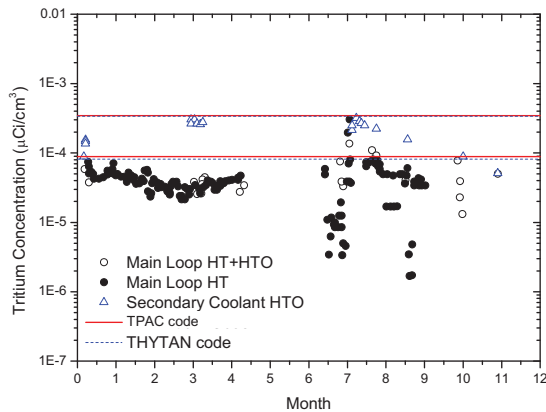

Fig. 5. Comparisons of tritium concentration in the Peach Bottom reactor between observed and computed ones.

IV. ANALYSES ON THE TRITIUM BEHAVIORS IN THE VHTR/HTSE SYSTEM

In order to understand tritium behaviors in the complex VHTR/HTSE systems, some preliminary modeling and simulations have been performed using the TPAC code. Fig. 6 illustrates a schematic of the reference system used in this study. This system is based on the indirect parallel configuration of a VHTR and a HTSE system, which was proposed by Oh et al. (2007) in their system optimization studies. In this configuration, the flow in the secondary coolant system is divided, with most of the flow going towards the PCU and the remainder going through a secondary heat exchanger (SHX) that directs heat towards the HTSE plant. The flow through the hot side of the SHX is then mixed with the flow from the PCU to feed the cold side of the intermediate heat exchanger (IHX). However, some of the flow is diverted away from the PCU, which acts to decrease the efficiency of the cycle. There are three coolant loops. The primary coolant system contains the nuclear reactor, the hot side of the IHX, and a compressor. The secondary coolant system contains the

cold side of the IHX, the hot side of the SHX, the PCU, and connecting piping, which is assumed to be short. The intermediate heat transport loop connects the secondary coolant system to the HTSE plant through several process heat exchangers (PHXs). For electrolysis, the steam is heated up to higher than 800 °C by the heat from SHX. The heated steam is converted into hydrogen and oxygen in the electrolyzer and discharged through the fuel and oxidizer outlet, respectively as shown in Fig. 7.
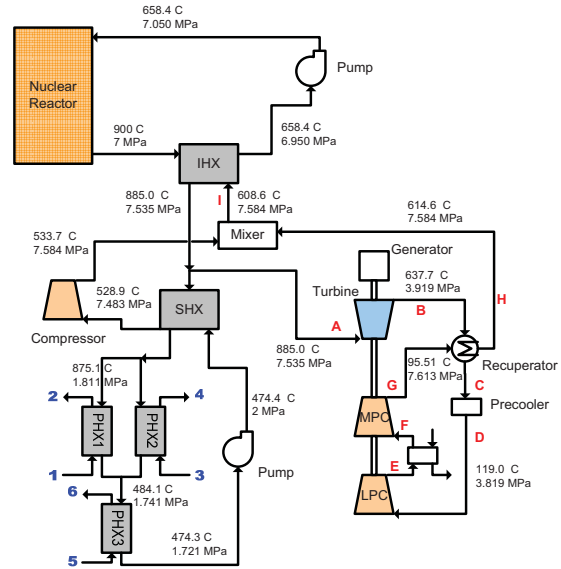

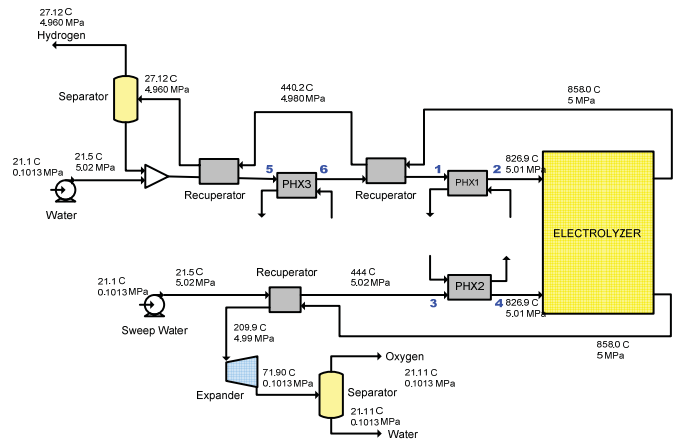Fig. 6. Reference system configuration (VHTR part).

Fig. 7. Reference system configuration (HTSE part).

Fig. 8 shows a snapshot of the TPAC model for the reference VHTR/HTSE system. In this modeling, the system has been divided into five separate loops; (1) primary side, (2) secondary side (PCU), (3) intermediate heat transfer loop (IHTL), (4) HTSE (water supplier), and (5) HTSE (purge flow). In this TPAC modeling, recycling and recuperating cycles have been eliminated from the HTSE system. This simplification does not change any main flow configurations and is expected to provide more
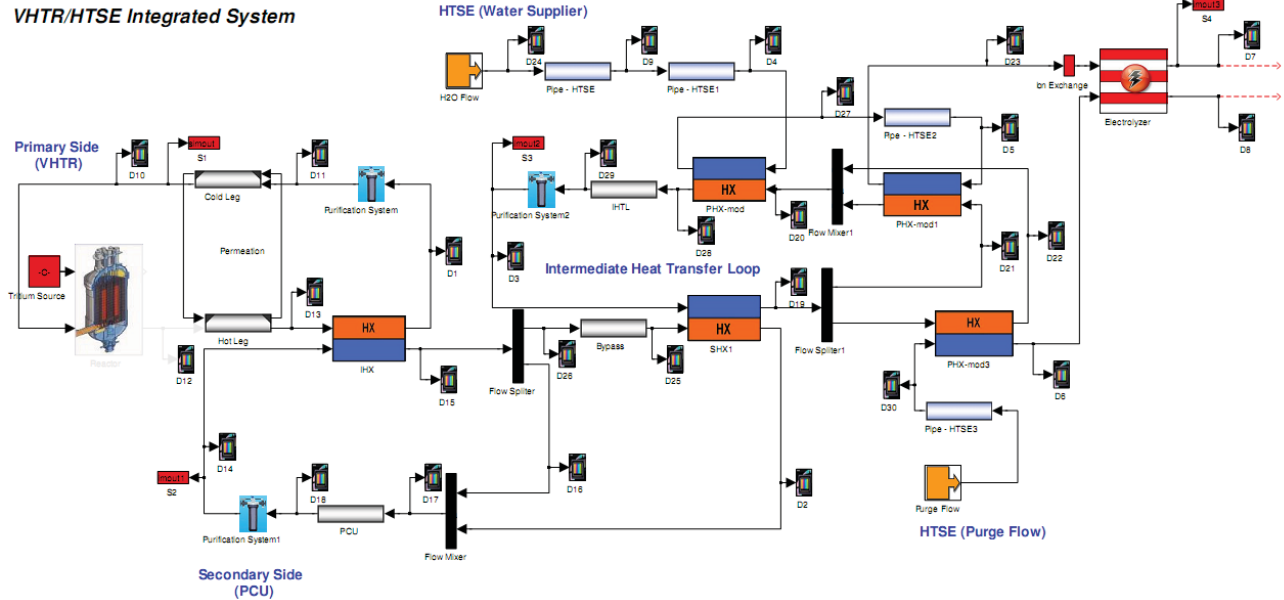
Fig. 8. TPAC modeling for VHTR/HTSE system (GUI).

conservative estimations with higher HT permeation out of the PHXs than the original one. In the TPAC model, the HTSE consists of the seven component blocks in the cathode side and four component blocks in the anode side.

TABLE III. Major input parameters.

Parameters	Value	Unit
Tritium Source (Release)	1.15E-11	m^3(STP)/s
Fraction of Mass Flow to Purification System (P-1st)	1.23E-4	#
Fraction of Mass Flow to Purification System (P-2nd)	1.22E-4	#
Fraction of Mass Flow to Purification System (P-3rd)	1.76E-4	#
Reaction Coefficient (IHX)	1.36e-10	#
Reaction Coefficient (SHX)	1.36e-10	#
Reaction Coefficient (PHX)	1.36e-10	#
Hx Thickness (IHX)	0.003	m
Hx Thickness (SHX)	0.003	m
Hx Thickness (PHX)	0.003	m
Average HX Temperature (IHX)	1060	K
Average HX Temperature (SHX)	999	K
Average HX Temperature (PHX)	981	K
HX activation energy (IHX)	5.35E+04	J/mol
HX activation energy (SHX)	5.35E+04	J/mol
HX activation energy (PHX)	5.35E+04	J/mol

Table III summarizes major input variables. The value of the tritium source was determined from previously reported tritium birth rates for the Peach Bottom reactor and the assumed release ratio. The reported tritium birth rate for this reactor is 2.90×10^{-11} Bq/y/MWt (Wichner and Dyer, 1979) and the assumed tritium release ratios are 0.1,

which is recommended in the TRITGO code that was originally developed at Oak Ridge National Laboratory to access tritium production and distribution in HTGRs (Compare 1974). By multiplying the tritium birth rates and the release ratios, the tritium release rates (tritium source) are estimated to be 1.15×10^{-11} m^3 (STP)/s. The mass flow into the purification system was assumed to be 20 %/h of the total helium inventory in each loop based on the Fort St. Vrain reactor design (Compere et al., 1974). According to those values, the fractions of mass flow into the purification systems are calculated to be 1.23×10^{-4}, 1.22×10^{-4}, and 1.76×10^{-4} in the primary, the secondary, and the IHTL sides, respectively. However, in the real VHTR/HTSE design, they can be adjusted to achieve acceptable gas chemistry in the system. The permeation coefficients and activation energies for permeation through the heat exchanger materials were determined based on the reported Alloy 800 material data. Yang et al. (1977) proposed 1.36×10^{-10} (m^3 (STP)/m/s/Pa$^{0.5}$) as the pre-exponential factor and 53.5×10^3 (J/mol) as the activation energy by analyzing Peach Bottom reactor. Heat exchanger designs including IHX, SHX, and PHXs were based on the compact heat exchanger type, and the design specifications were taken from the report published by Oh et al. (2008). The wall thicknesses of the heat exchanger were assumed to be 3 mm. Hydrogen concentration in the He coolant was assumed to be 10 ppmv (GA, 2009). Other chemical impurities such as humidity and hydrocarbon were not considered in this modeling.

Fig. 9 plots the time variations of the tritium concentrations in the product hydrogen. As shown in this figure, the tritium concentration in the hydrogen product are well converged to a steady state conditions after about 1×10^5 sec. In this simulation, the estimated tritium

concentration in the hydrogen product is 2.16×10^{-5} ppb (1.04×10^{-3} Bq/cm^3(STP)). Fig. 9 also compares the tritium concentrations predicted in the product hydrogen with the tritium effluent limit (3.7×10^{-3} Bq/cm^3(STP)). As shown in this figure, the tritium concentration in the hydrogen product was estimated to be lower than the effluent limit by factor 3 to 4. Based on the improved system design and tritium protection methods, the tritium concentration in the product hydrogen can be even much lower than this level. According to the recent extensive reviews on the tritium barrier studies (Sherman and Adams, 2008), oxide layers produced in-situ or ceramic coating layers can significantly reduce tritium permeation through the heat exchanger surface by a factor of 10 to over 10,000. Therefore, the tritium concentration in the hydrogen product can be even much less than that (1.04×10^{-3} Bq/cm^3(STP)) reported in this paper in the future by incorporating these novel methods into the actual heat exchanger design and fabrication.
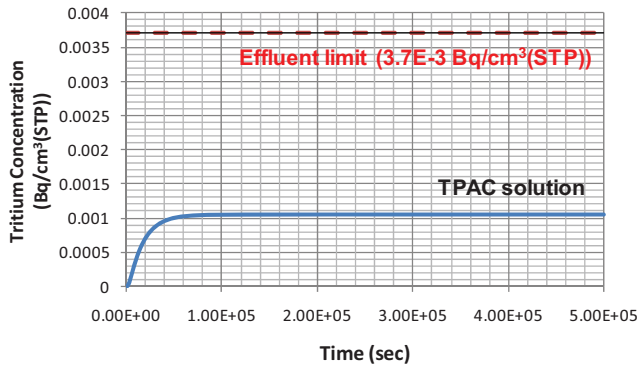

Fig. 9. TPAC calculations for tritium concentrations in the product hydrogen.

Fig. 10 illustrates the estimated tritium distributions in the VHTR/HTSE system for the reference conditions. As shown in this figure, about 54.2 % of the released tritium from the core is removed by the purification system installed in the primary side. About 45.7 % of the tritium is transferred to the secondary side through the heat exchanger wall. Only 0.1 % of tritium is released to the containment (or confinement) by helium leakage. In the secondary side, majority of the tritium (34.5%) is removed by the purification system. 11.3 % of the tritium released from the core is transferred to the intermediate heat transfer loop through the heat exchanger. In the intermediate loop, 1.82 % of the tritium released from the core is removed by the purification system. About 3.52 % of the tritium released from the core is transferred to the product hydrogen. According to the calculation results, the purification systems in the primary and secondary system looks the most important for tritium transfer removal and distribution in the system. The effect of leakage out of the system looks negligible, which is less than 0.2 % in total. However, this distribution can be quite differed from the

other system designs, configurations and conditions. Therefore, careful interpretations are required for general extension of this result to other systems.
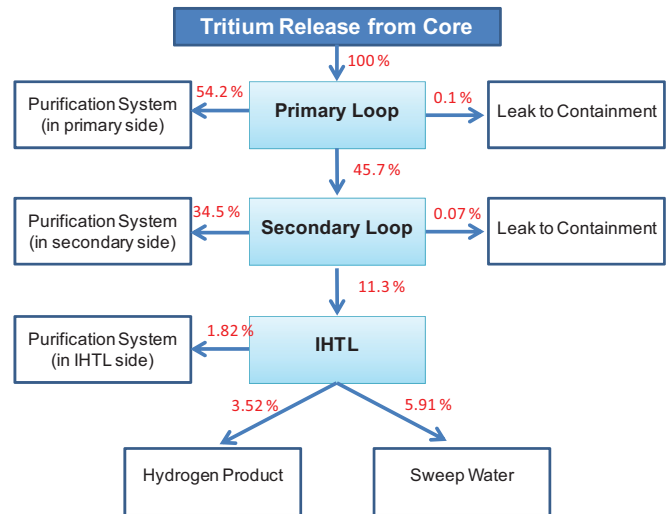

Fig. 10. Tritium distribution in the reference VHTR/HTSE system.

IV. SUMMARY AND CONCLUSIONS

A tritium analysis code named by TPAC has been developed by INL using MATLAB SIMULINK for analyses of tritium behaviors in the VHTR systems. This code is based on the mass and species conservation laws of the tritium or hydrogen containing molecules such as HT, H_2, and HTO for general tritium permeation, and H_2O, H_2SO_4, $HTSO_4$, TI, specifically for the SI process application. Isotope exchange reactions for other industry application can be also implemented in the near future. Various tritium birth models have been implemented into the code. The models are ternary fission and neutron reactions with impurities (^6Li, ^7Li, ^{10}B, ^3He) in the core structures and the coolant. An Arrehius type equation form was used for tritium permeation through the heat exchanger walls, vessel walls, and pipe walls. Leakage model, purification system model, electrolyzer model, and ion-exchange models have been also implemented in the code.

Verification of the TPAC code has been performed by analytical solutions and experimental data based on the Peach Bottom reactor. Various separate models have been taken into considerations for verification (e.g. tritium birth models, leakage model, permeation model, purification system model, ion-exchange model, and etc.). As a result, the TPAC calculation results showed very good agreement with the analytical and experimental data. The integrated system model for the Peach Bottom reactor has been analyzed by the TPAC code and compared with the

reported data for verifying total integration and performance of the code. The TPAC code showed reasonable agreement with the reported data in the Peach Bottom reactor giving rather conservative prediction. However, the quality of the measured data was not good enough for validation and the input conditions seems to have large uncertainties. For validation of the code, more reliable experimental or plant operation data are strongly recommended with well-defined input parameters.

Tritium concentration and distribution in the VHTR and industry process application side depend on the system configuration, operating conditions, heat exchanger surface area, wall thickness, and the concentration difference between the hot side and cold side of the heat exchanger. In this paper, some preliminary analyses on the tritium behaviors in the VHTR/HTSE system have been carried out by the verified TPAC code. The reference reactor configurations and operating conditions were taken from the indirect parallel configuration proposed by Oh et al. (2007). Geometrical information for the reactor has been determined by the current VHTR pre-conceptual designs and some assumptions. Heat exchanger designs including IHX, SHX, and PHXs were based on the PCHE type heat exchangers and the design specifications were taken from the report published. Information related to the tritium birth, purification, and leakages were based on various previous literatures (Richards 2006, GA 2006, Ohashi and Sherman 2006, Wichner and Dyer 1979). The following summarizes the notable results and discussions on this analysis.

•The tritium concentration in the product hydrogen was estimated to be about 0.001 Bq/cm^3[STP], which is lower than the regulatory tritium effluent limit (=0.0037 Bq/cm^3[STP]) by a factor of 3 to 4. This tritium concentration level can be even reduced by improved system designs and tritium permeation barrier methods.

•According to the preliminary analyses, about 54.2 % of the released tritium from the core is removed by the purification system installed in the primary side. About 45.7 % of the tritium is transferred to the secondary side through the heat exchanger wall. Only 0.1 % of tritium is released to the containment (or confinement) by helium leakage. In the secondary side, majority of the tritium (34.5%) is removed by the purification system. 11.3 % of the tritium released from the core is transferred to the intermediate heat transfer loop through the heat exchanger. In the intermediate loop, 1.82 % of the tritium released from the core is removed by the purification system. About 3.52 % of the tritium released from the core is transferred to the product hydrogen. The effect of leakage out of the system looks negligible, which is less than 0.2 % in total.

NOMENCLATURE

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)]

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]

K = fission rate per thermal megawatt [fission/MW/s]

$k_{p,T}$ = permeability of tritium [m^3 (STP)/m/s/Pa$^{0.5}$]

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]

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

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

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

$P_{Total,l}$ = 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].

$S_{i,j}$ = 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]

T_0 = standard temperature (273.15 K)

V_j = the 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]

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