DEVELOPMENT AND VERIFICATION OF BEHAVIOR OF TRITIUM ANALYTIC CODE (BOTANIC)

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ABSTRACT

Very High Temperature Reactor is known for its capability to provide process heat to various industrial processes; ammonia synthesis, desalination process and hydrogen production. This is possible due to its high operation temperature which satisfies temperature constraints of most of the industrial processes. This characteristic has been known as the strength of the VHTR. However, there is a potential safety issue related to tritium.
Tritium is a radioactive hydrogen isotope which is produced in ternary fission reactions and neutron reactions in nuclear reactor systems. It typically exists in tritium gas form (HT) and has a habit of easily permeating through metals. The permeation rate of tritium increases with increasing temperature and this is why tritium is treated more severely in Very High Temperature Reactors (VHTRs). It possesses a high operation temperature and additionally it provides heat to other industrial processes which opens a possibility of tritium permeating to the industrial process. Thus, it is vital to understand the tritium behavior in the VHTR systems and the integrated industrial processes.
In this study, a Behavior of Tritium Analytic code, an analytical tool which is capable of analyzing tritium behavior in both component level and system level, is developed using a chemical process code gPROMS. For accuracy, a very detailed diffusion and trapping based permeation model has been implemented. The developed BOTANIC code was then further verified using the analytical solutions and the benchmark codes such as Tritium Permeation Analysis Code (TPAC) and COMSOL Multiphysics code.

KEYWORDS
Tritium, VHTR, Permeation, PCHE

1. INTRODUCTION

Very High Temperature Reactor (VHTR) is one of the six Generation IV reactor concepts suggested. It is a gas cooled graphite moderated reactor with a once-through uranium fuel cycle. The concept was first suggested in the early 1950's and research on the reactor concept has been conducted ever since. As can be expected from its name, VHTR is known to operate at a very high temperature, up to 1000 ˚C. And due to its high operation temperature, high overall efficiency is achievable. Furthermore, as the operation temperature of the VHTR satisfies temperature constraints of various industrial processes, providing process heat to industrial process is viable. These two features, high overall efficiency and capability to provide process heat to various industrial processes have allowed VHTR to gain worldwide attention. But

at the same time these features are the reasons why tritium is treated more severely in VHTRs than in other reactor types.

Tritium is a hydrogen isotope with two more neutrons than the typical hydrogen. Due to its structure, it is unstable and thus radioactive. Tritium goes through beta decay and has a half-life of 12.32 years. It typically exists in a hydrogen-tritium (HT) gas form or tritiated water (HTO) form. Tritium is very small and light and has an instinct of easily permeating through solid metals. And this permeability is greatly affected by the temperature. Permeability of tritium increases with increasing temperature and since VHTR operation temperature is relatively high, it is likely that tritium permeation will occur more and faster. Furthermore, since VHTR is integrated to an industrial process, there is a possibility of tritium permeating through the integration loop and into the industrial process or even into the final product of the industrial process. In other words, due to its high operation temperature and the possibility of the integration to other industrial process, tritium is a critical safety issue in VHTR.

Natural occurrence of tritium in the environment is extremely rare. Only 10-16 % of the naturally occurring hydrogen is made up of tritium. It goes through beta decay and releases at maximum 18.6 keV of energy during this process. The low-energy beta particle emitted by tritium has a maximum range in water or tissue of 6 mm, thus, it can be said as non-hazardous externally [1]. However, beta radiation is a type of ionizing radiation which can cause cell damage and further cause cancer or genetic disorder when exposed inside the body. Given its low energy beta emission and the short range in air, tritium poses a health risk only when ingested, inhaled or absorbed through skin. The biological half-life of tritium depends on the form and the method of intake. Only about 0.004 percent of inhaled tritium gas is retained more than a minute or so, the rest is eliminated almost immediately through respiration. When tritium is ingested in tritiated water form it immediately mixes with the body fluids and has a half-life of 10 days [2]. And as tritiated water act as water in the body, tritium is distributed through all biological fluids within 1 to 2 hours. In other words, tritium is hazardous when ingested in the body. And as a result, regulatory limits have been set for tritium in drinking water. The WHO limits tritium level in drinking water to 10,000 Bq/L.

Tritium is generated and lost through various phenomena in a nuclear reactor system. The processes include tritium production, leakage, purification and permeation. Most of the tritium is generated in the reactor core and majority of tritium is generated through ternary fission. Tritium is also generated through neutron reactions within the core. Most of the tritium generated in the core is released in the coolant and removed from the system through purification. Portion of tritium is leaked out of the system and some permeate into secondary or even to the integration loop.

As seen here tritium is a critical safety issue and involves complex mechanisms in a nuclear reactor system thus, the analysis of tritium behaviour in the system is vital. Accordingly efforts have been made in this extent. Researches regarding the tritium analysis in the VHTR have been conducted by many research groups [3][4][5]. Codes such as Tritium and Hydrogen Transportation Analysis Code (THYTAN), Tritium Permeation Analysis Code (TPAC), Tritium Migration Analysis Program (TMAP) have been developed [3][4][5]. However, the existing codes such as THYTAN, TPAC and TMAP have certain limitations. The codes focus on specific functions. For example, THYTAN and TPAC focus on the tritium analysis in system level and TMAP has the capability to analyse tritium in a component level in a more detailed manner. And all of these codes are incapable of process modelling. And this is because the existing codes use the diluted assumption and only solves tritium mass conservation equation. Due to these limitations, in order to perform an analysis on a system 2 to 3 codes are required which can possibly cause inconvenience and uncertainties. This is the main reason for developing the Behavior of Tritium Analytic Code (BOTANIC).

In this study, tritium analysis code is developed based on a chemical process code called gPROMS [6]. BOTANIC possesses several distinctive features such as non-diluted assumption and capacity to perform system dynamics calculation including mass, momentum and energy conservations with detailed permeation model and limited multidimensional capability. The developed code was then verified using the analytical solution and TPAC calculation results generated based on the peach bottom reactor core II operation data.

2. TRITIUM BEHVIOUR

2.1. Tritium Pathway

Tritium is generated through various reactions. In VHTR, tritium is mainly produced as a byproduct in ternary fission reactions. Tritium is also generated in neutron capture reactions with the core and coolant materials such as ^{6}Li, ^{3}He, ^{10}B and ^{7}Li. ^{6}Li and ^{7}Li are impurities in graphite core components such as sleeve, spine, reflector and fuel matrix. ^{10}B exists in control rods, burnable poisons and reflectors. And ^{3}He is a helium isotope which exists as an impurity in the coolant.

The produced tritium is transported and lost through various processes in the reactor system as shown in figure 1. A portion of the generated tritium is retained in the solid structures and the rest is released to the coolant. Most of the tritium circulating in the primary coolant system is removed in the purification system. Some of it escapes to the environment through leakage and some transport to the secondary system through permeation. The tritium in the secondary system is lost in a similar manner as in the primary system. As some of tritium transported across the heat exchanger from primary to secondary loop, a portion of tritium transports from the secondary system to the industrial process system through the process heat transfer heat exchanger.

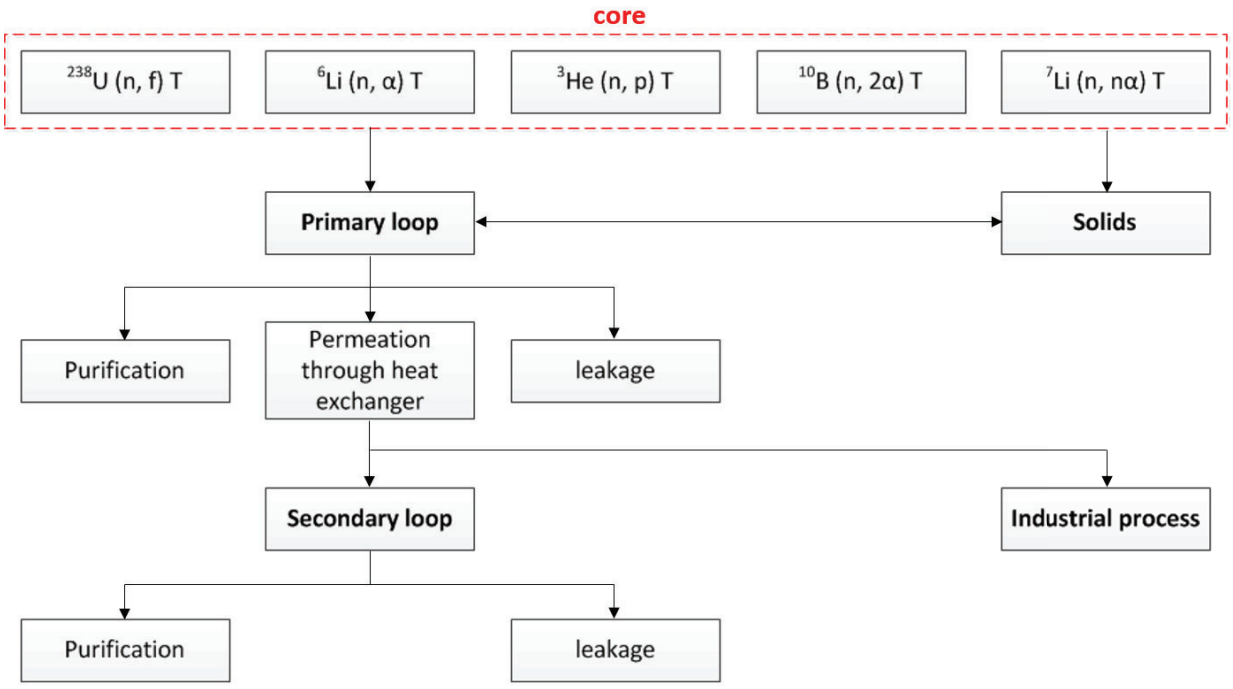

Figure 1. Tritium pathway in VHTR system.

2.2. Tritium Generation

As mentioned, tritium is generated through various reactions in the reactor core. The main tritium birth mechanism is ternary fission of the fuel (uranium-233, uranium-235, plutonium-239 and plutonium-241) in the core due to thermal neutrons. Tritium is also produced in neutron capture reactions in the core. The models implemented in the BOTANIC code can be seen in Table 1.

Table 1. Tritium generation model [4]

Model description	Analytical model
Ternary fission model	$\frac{d(N_{T(Ter)})}{dt} = K \cdot P \cdot Y - \lambda \cdot N_{T(Ter)}$ (1)
Lithium-6 model	$\frac{d(N_{Li6})}{dt} = -\phi_{th} \cdot \sigma_{Li6T} \cdot N_{Li6}$ (2) $\frac{d(N_{T(Li6)})}{dt} = \phi_{th} \cdot \sigma_{Li6T} \cdot N_{Li6} - \lambda \cdot N_{T(Li6)}$ (3)
Helium-3 model	$\frac{d(N_{He3})}{dt} = f \cdot N^{\circ}_{He3} - f \cdot N_{He3} - \phi_{He} \cdot \sigma_{He3T} \cdot N_{He3}$ (4) $\frac{d(N_{T(He3)})}{dt} = \phi_{He} \cdot \sigma_{He3T} \cdot N_{He3} - \lambda \cdot N_{T(He3)}$ (5) $\phi_{He} = \frac{W_{core}}{W_{total}} \cdot \phi_{th}$ (6)
Boron-10 model	$\frac{d(N_{B10})}{dt} = -(\phi_{th} \cdot \sigma_{B10Li7} + \phi_f \cdot \sigma_{B10T}) \cdot N_{B10}$ (7) $\frac{d(N_{Li7(B10)})}{dt} = \phi_{th} \cdot \sigma_{B10Li7} \cdot N_{B10} - \phi_f \cdot \sigma_{Li7T} \cdot N_{Li7(B10)}$ (8) $\frac{d(N_{T(B10)})}{dt} = \phi_f \cdot \sigma_{Li7T} \cdot N_{Li7(B10)} + \phi_f \cdot \sigma_{B10T} \cdot N_{B10} - \lambda \cdot N_{T(B10)}$ (9)
Lithium-7 model	$\frac{d(N_{Li7})}{dt} = -\phi_f \cdot \sigma_{Li7H3} \cdot N_{Li7}$ (10) $\frac{d(N_{T(Li7)})}{dt} = \phi_f \cdot \sigma_{Li7T} \cdot N_{Li7} - \lambda \cdot N_{T(Li7)}$ (11)

Lithium-6 and lithium-7 are impurities in the core graphite structure; sleeve, spine, reflector and fuel matrix. Basically, the thermal neutron from the core react with the lithium-6 in the graphite structure. Some of the tritium atoms produced in the neutron reaction are captured within the solid structure and some diffuse through the solid structure. Once the tritium atom reaches the surface of the structure it tends to bind with hydrogen and gets released into the helium coolant. The reaction mechanisms of lithium-7, boron-10 and helium-3 are similar to that of the lithium-6 neutron capture reaction. The main difference lies in the location of the reaction occurrence and the release mechanism. Helium-3 neutron reaction occurs within the coolant as the helium isotope is located inside the coolant. And since the reaction occurs within the coolant the generated tritium directly binds with the hydrogen in the coolant to form HT gas and distributes inside the system.

2.3. Tritium Leakage

In the VHTR, leakage of coolant occurs through defects or micro-gaps between components such as pipe, flanges, etc. As there is a certain concentration of tritium present in the coolant, tritium is lost with the bulk coolant. The leak rate, S_{leak} of chemical i, is calculated using equation 12. MT represents the total mass holdup of the node, L_R is the leakage rate. And x_i is the mass fraction of chemical i, in this case tritium, in the node.

$$S_{leak,i} = MT \cdot L_R \cdot x_i \tag{12}$$

2.4. Tritium Purification

Most of the tritium in primary and secondary loops is removed in the purification system. The removal rate of tritium through purification is expressed in equation 13. Where $F_{PF,He}$ is the flow rate to the purification system and η_i is the fractional purification efficiency of tritium in the purification system. The purification efficiency of tritium typically exceeds 95 %.

$$S_{PF,i,j} = F_{PF,He} \cdot \eta_i \cdot x_i \tag{13}$$

2.5. Tritium Permeation

Permeation is the core tritium transport mechanism in most of the nuclear reactor systems. Permeation phenomena has been noted as the most important activity through sensitivity analyses [7]. Since permeation is the key mechanism in tritium analysis, the BOTANIC code adopts a detailed permeation model unlike other existing tritium analysis codes in addition to the simple correlation based permeation model. For convenience BOTANIC provides a variety of permeation models including the simple correlation based permeation model, equilibrium permeation model and non-equilibrium permeation model.

Tritium permeation mechanism is a quite complex process. It involves a complicated reaction mechanism on the surface of a metal and also within the solid structure. On the solid surface recombination and dissociation occurs as shown in figure 2. And within the solid structure trapping, release and diffusion governs the atomic movement.

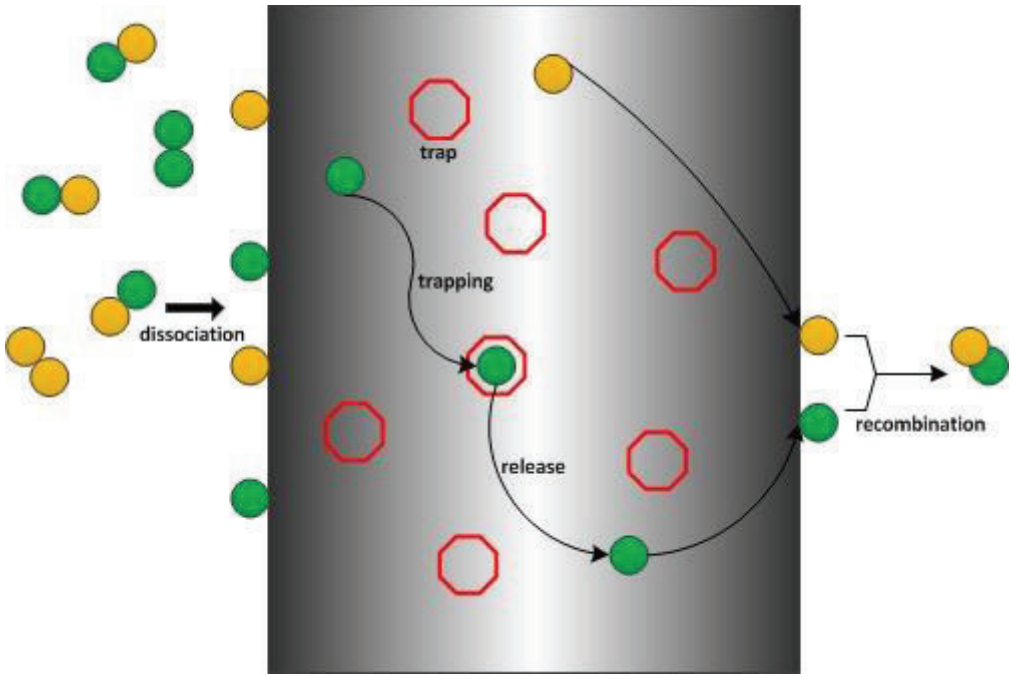

Figure 2 Tritium permeation mechanism

The simple correlation based permeation model basically ignores these mechanisms and is built based on Sievert's law. This model is widely used. Most of the existing tritium analysis codes including THYTAN uses this equation 14 [4].

$$S_{m,permeation,i} = \frac{1}{1000} M_i \cdot k_p \cdot \frac{A}{l}\left(\sqrt{P \cdot y_i} - \sqrt{P \cdot y_{i,o}}\right) \tag{14}$$

However, considering the importance of the phenomena, a more detailed model which considers the surface mechanisms and the mechanisms inside the structure is adopted in the BOTANIC code; equilibrium model and non-equilibrium model. The distinction between the equilibrium model and non-equilibrium model is the surface mechanism assumption. If the dissociation rate and recombination rate are assumed identical, the equilibrium model is used. When the rates are considered separately, the non-equilibrium model applies.

In the case of equilibrium model, the surface concentration is calculated using the Sievert's law in equation 15. The movement across the solid structure is calculated using equation 16. The equation includes the trapping term, diffusion term, release term and the decay term. In the case of the non-equilibrium model, the surface concentration is calculated using equation 17. The model considers the atomic flux to the surface and the diffusion rate from the surface. For the movement across the solid structure, equation 16 is used.

$$C_i{}^2 = K_i \cdot P_m \tag{15}$$

$$\frac{dC_i}{dt} = -\nabla^2 C_i + S_i - \left\{\frac{\alpha_{t_i} C_{tk}^e}{N} C_i - (\alpha_{rk} + v_i) C_i^{tk}\right\} \tag{16}$$

$$\sum_j a_{m_s}(K_{d_m} P_m - \sum_{i,j} K_{r_m} C_i{}^2) + D\nabla C_i = 0 \tag{17}$$

3. DEVELOPMENT OF BOTANIC

3.1. Overview

The BOTANIC code is developed based on the chemical process code called gPROMS. gPROMS is a platform for modeling process industries. It provides modelling interface, user interface creation and Process Model Library. The Process Model Library is composed of commonly used components such as pipe, pump, heat exchanger, etc. The BOTANIC code is developed by implementing necessary tritium models in the associated component models. As the BOTANIC code is developed based on the chemical process code it is capable of analyzing chemical process, process modelling and multi-dimensional tritium analysis.

One of the most distinctive characteristics of BOTANIC is that it does not use a diluted assumption. Diluted assumption is an assumption used when one assumes that tritium level is extremely low, low enough to neglect the effect of tritium generation or loss in the overall physical property. Most of the existing tritium codes use the diluted assumption which results in limitation in the analysis; only low level situations can be analyzed and possibly accuracy. As BOTANIC reflects the effect of tritium generated or lost in the total system properties, it can be used to analyze high or low tritium situations. In other words, although BOTANIC is originally aimed to analyze VHTR systems, it has the capacity to analyze fusion reactor systems. This flexibility of BOTANIC is a great possibility and strength.

Another notable feature of BOTANIC is that it can solve both the lumped and multi-dimensional model. BOTANIC provides complex components such as heat exchanger and reactor as distributed model while other simple components in lumped model. By composing BOTANIC in an efficient manner, the user can achieve a short calculation time and accurate calculation result at the same time.

Lastly, BOTANIC solves mass, energy and momentum conservation equations. It is briefly mentioned that BOTANIC is capable of analyzing system dynamics. This is only possible as it solves the three conservation equations. The code has the capability to calculate properties such as temperature, mass, flow rate, and physical properties only because it solves the three conservation equations. In the conventional tritium codes, only mass conservation equation regarding tritium was solved thus, additional calculations had to be conducted in order to gain the necessary system properties.

3.2. BOTANIC Structure

The components in the BOTANIC code can categorized as node component and junction component. Typically node components solve mass and energy conservation equations and junction components solve momentum conservation equation. Associated tritium models are inserted in each component. The code structure can be seen in Figure 3. As seen in figure 3, related tritium models are inserted in the node component and by solving the mass and momentum equations in the node component together with tritium model, variables such as mass fraction, pressure and temperature are calculated in the node component. The information is then transferred to the junction component where momentum equation is solved as seen in the figure. Calculated flow rate is then delivered to the adjacent component together with the information transferred from the previous node component.

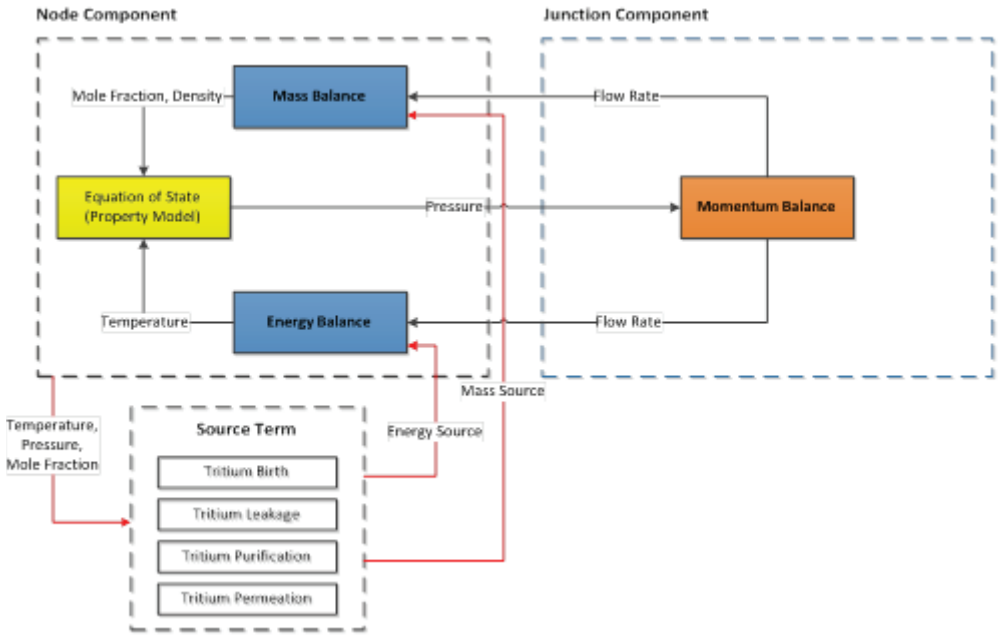

Figure 3. BOTANIC code structure

4. VERIFICATION OF BOTANIC

4.1. Overview of the Verification Process

BOTANIC is verified in two levels; verification of the individual tritium models and verification of the component. The individual tritium models are verified using the analytic solution and the benchmark code calculation result. Then components containing the tritium models are verified in terms of integration; whether the models are well implemented and connected within the component model.

4.2. Verification of Tritium Generation Model

Tritium birth models were verified using the Peach Bottom Core 2 data. The conditions from the Peach Bottom Core 2 were used and the calculation results were then compared with the reported data from the Peach Bottom. The calculation time was set as 1,550 days [8], the operation time of Peach Bottom Core 2.

Calculation results of BOTANIC showed very good agreement with the Peach Bottom values. In the ternary fission model calculation the reported value, analytical solution and the calculation result of BOTANIC were identical as 4.43×10^{13}. Verification result of ^{6}Li model showed very good agreement with the analytical solution as shown in table 2.

Table 2. Verification result of Lithium-6 model

Graphite component	Analytical solution (Bq)	BOTANIC (Bq)
Sleeve	5.12×10^{11}	5.12×10^{11}
Spine	3.78×10^{10}	3.78×10^{10}
Removal radial reflector	5.76×10^{11}	5.53×10^{11}
Permanent radial reflector	6.72×10^{11}	5.69×10^{11}
Axial reflector	3.42×10^{11}	3.42×10^{11}
Fuel matrix	5.68×10^{11}	5.67×10^{11}

The helium model verification result showed very good agreement with one of the benchmark code called Tritium Permeation Analyses Code (TPAC) as seen in table 3. And the ^{10}B model calculation result in BOTANIC was identical to that of the analytical solution; 3.19×10^{12}.

Table 3. Verification result of helium-3 model

Region	TPAC (Bq)	BOTANIC (Bq)
Coolant	1.10×10^{12}	1.10×10^{12}
Sleeve Graphite	1.58×10^{11}	1.58×10^{11}
Removal Radial Reflector	3.87×10^{11}	3.87×10^{11}
Permanent Radial Reflector	5.33×10^{11}	5.33×10^{11}
Axial Reflector	8.98×10^{10}	8.98×10^{10}

4.3. Verification of Tritium Leakage Model

The BOTANIC tritium leakage model was verified using the benchmark code TPAC. Figure 4 shows the flowsheet modelling of the leakage verification case. Very simple case where leakage occurs from the primary loop to the containment was used for verification. The containment vessel temperature was set as 323 K and the tritium concentration in the primary loop was 1.98×10^3 ppb under the reported average temperature of 809 K and 23 atm in the primary coolant [8]. The initial tritium concentration in the containment building was assumed to be 0. The leak rate from the primary loop to the coolant and the leak rate from the containment to the atmosphere was assumed as 4.15×10^{-4} 1/h. The calculated tritium level in the containment component using TPAC and BOTANIC was compared in figure 5. And as seen in the figure the calculation results show very good agreement with each other.

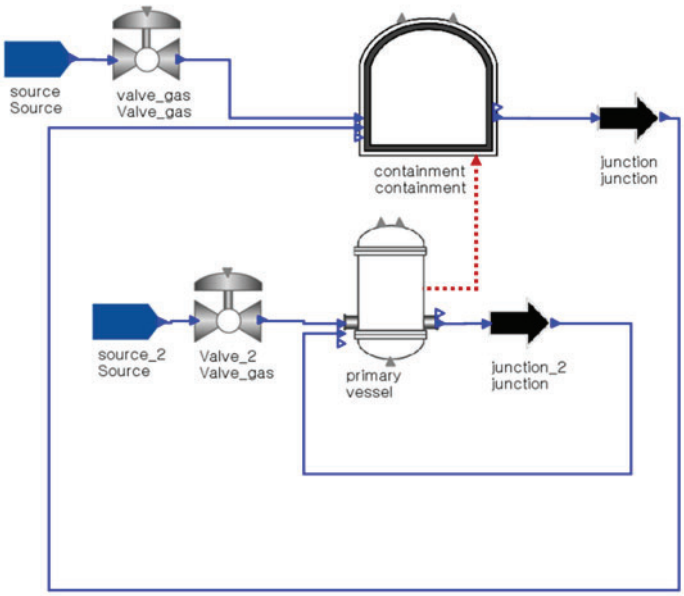

Figure 4. Leakage flowsheet model

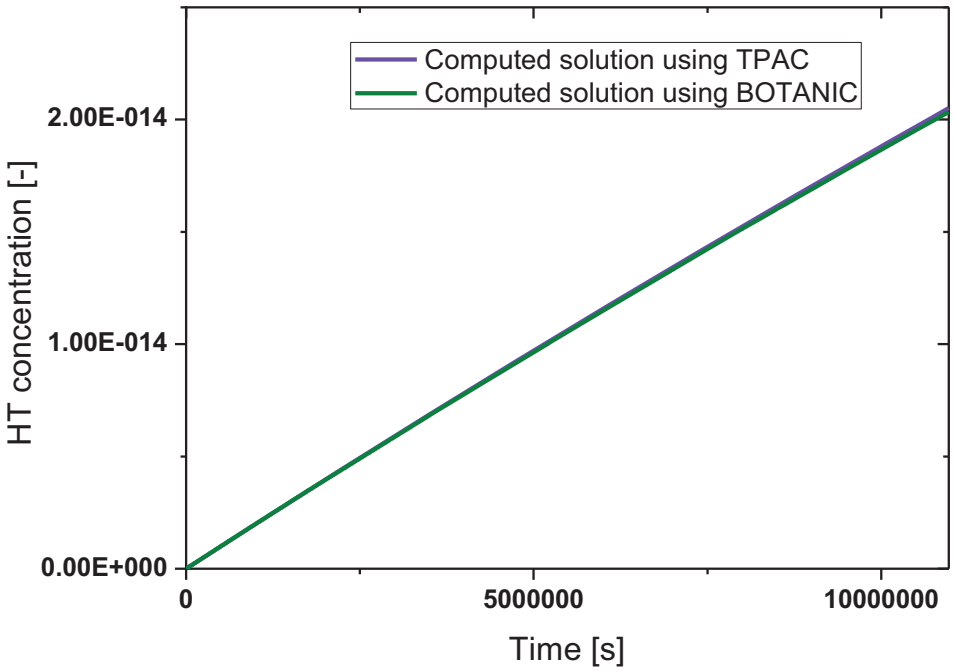

Figure 5. Leakage verification result

4.4. Verification of Tritium Purification Model

Purification model was verified using a simple case. The purification system inventory was assumed as 1 m^3 and the purification efficiency of HT was set as 90 %. The helium flow rate to the purification system was 1.0×10^{-3} m^3/s. And the initial concentration of HT was assumed to be 1.0×10^{-9} [8]. The flowsheet modelling of purification verification case is as in figure 6. And as can be seen in figure 7, the purification calculation result show very good agreement with the analytical solution.

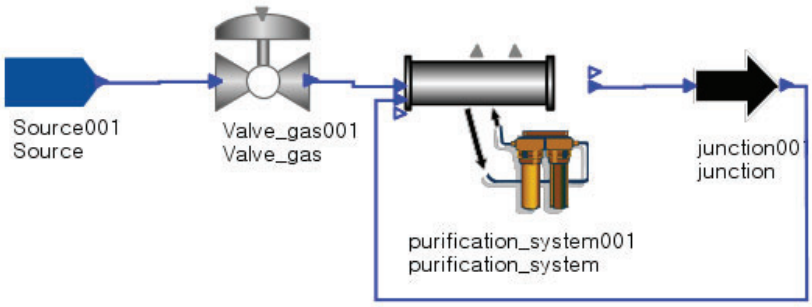

Figure 6. Purification flowsheet model

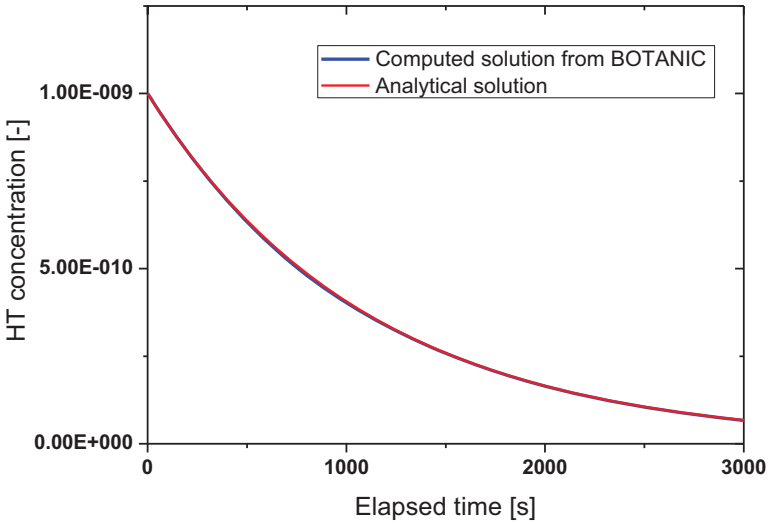

Figure 7. Verification result of the purification model

4.5. Verification of the Permeation Model

The simple correlation permeation model was verified using the analytic solution. For the verification process, the heat exchanger component model was used. The permeation rate calculated using BOTANIC code and the analytical solution were calculated to be identical as seen in Table 4.

Table 4. Verification result of permeation model

Parameters	Unit	Analytic	BOTANIC
Permeability		1.59×10^{-17}	1.58×10^{-17}
Permation rate	m^3/s	9.48×10^{-13}	9.31×10^{-13}
Perm_rate	kg/s	1.27×10^{-13}	1.25×10^{-13}

The Benchmark codes do not provide the multidimensional permeation model and the detailed permeation model. Thus, the multidimensional equilibrium model and the non-equilibrium model had to be verified using the multi-physics code, COMSOL. Firstly, the surface models were verified. Surface concentration was calculated using the Sievert's law as 0.009487 in both BOTANIC and COMSOL when the equilibrium constant was assumed as 0.003, pressure 100000 Pa and mass fraction 0.0001. The wall thickness was assumed as 2 m and the wall length as 4 m. Wall temperature was calculated using a linear equation and the diffusivity was calculated using the Arrhenius equation. Diffusivity activation energy was given as 3.74×10^4. And the jump constant was assumed as 4.7×10^{-10}, trapping energy as 490. Calculation results of BOTANIC and COMSOL is shown in figure 8 and 9. Figure 8 is the concentration of tritium at the center of the wall. Figure 9 is the tritium distribution across the solid structure at 1000 s. As seen in the figure the calculation results seem to be in agreement.

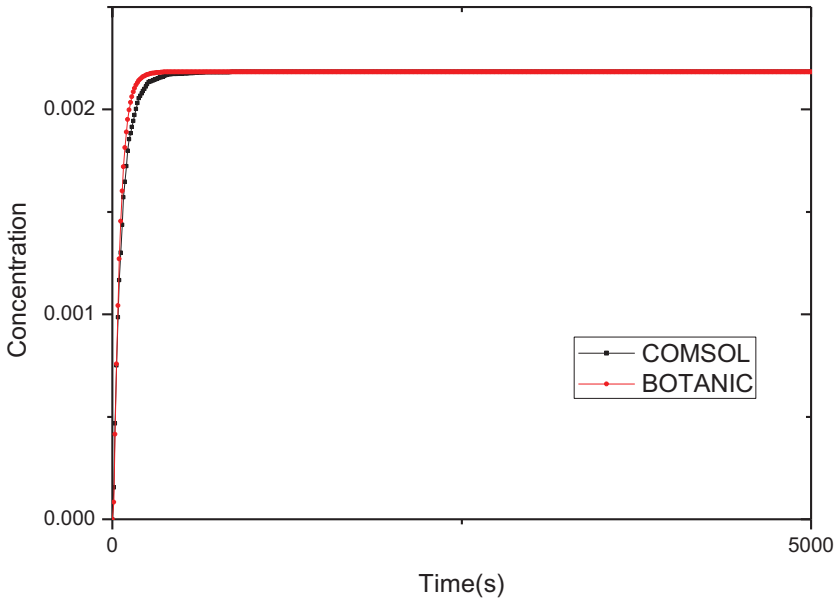

Figure 8 Tritium concentration at the wall center

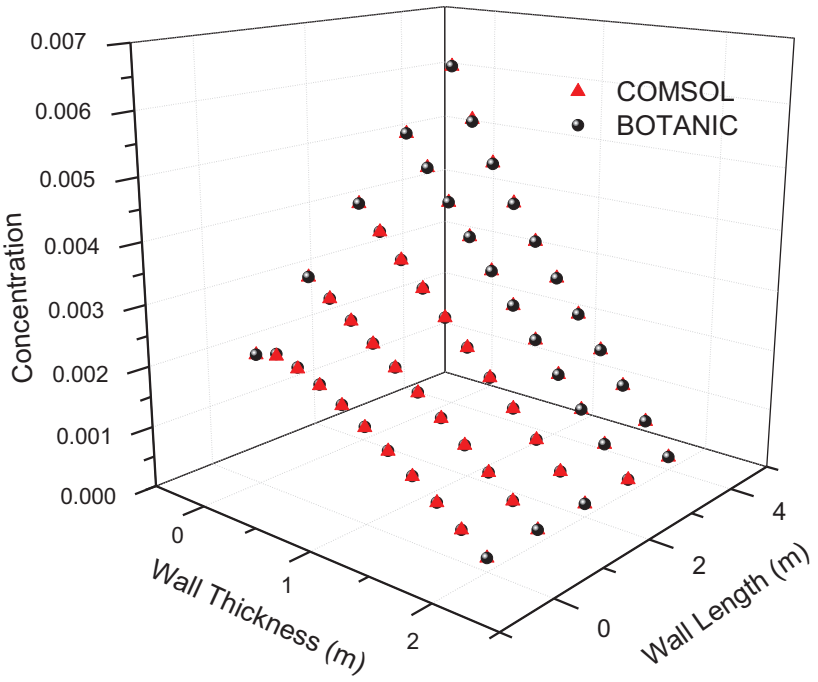

Figure 9 Tritium concentration distribution across the solid structure at t=1000 s

5. CONCLUSION

In this study, the Behavior of Tritium Analytic Code, BOTANIC, has been developed using a chemical process code called gPROMS. The code has several distinctive advantages including non-dilution assumption, flexible applications and adoption of multidimensional detailed permeation model. Due to these features the BOTANIC code has the capability to analyze a wide range of tritium level systems and furthermore it maximizes the user's convenience and accuracy as it provides multidimensional model and process modeling. The developed code was successfully verified using analytical solution and the benchmark code calculation result. The BOTANIC calculation results showed very good agreement with the analytical solutions and the calculation results of TPAC and COMSOL.

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