Development and Verification of Behavior of Tritium Analytic Code (BOTANIC)

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1. Introduction

Tritium is one of the key issues in the fission reactor systems, especially in the Very High Temperature Reactor (VHTR) system. Tritium is a radioactive hydrogen isotope with a half-life of 12.32 years [1]. 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 VHTRs. VHTR, one of the Generation IV reactor concepts, has a relatively high operation temperature and is usually suggested as a heat source for many industrial processes, including hydrogen production process. Thus, it is vital to trace tritium behavior in the VHTR system and the potential permeation rate to the industrial process. In other words, tritium is a crucial issue in terms of safety in the fission reactor system. Therefore, it is necessary to understand the behavior of tritium and the development of the tool to enable this is vital. In this study, a Behavior of Tritium Analytic Code (BOTANIC) an analytic tool which is capable of analyzing tritium behavior is developed using a chemical process code called gPROMS [2]. BOTANIC was then further verified using the analytic solutions and benchmark codes such as Tritium Permeation Analysis Code (TPAC) and COMSOL [3][4].

2. Tritium behavior

2.1 Tritium Behavior

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

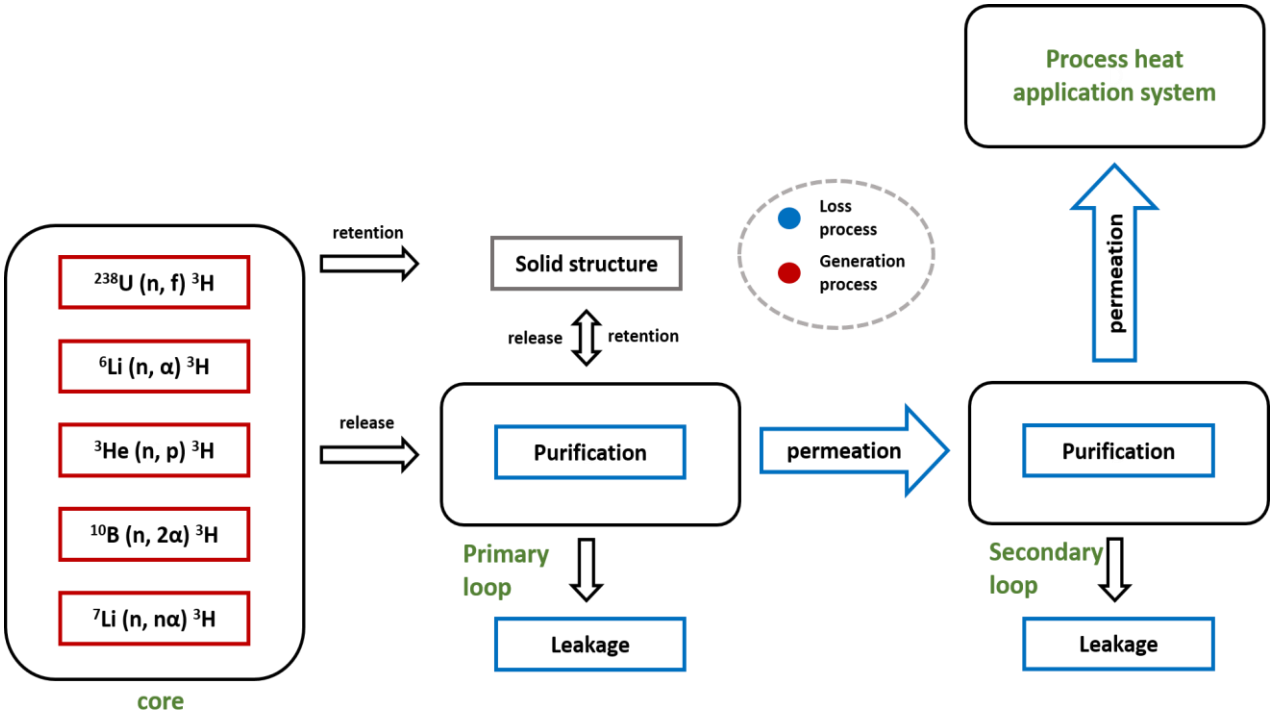

Figure 1 Tritium behavior in the VHTR system [5]

secondary system to the industrial process system through the process heat transfer heat exchanger.

2.2 Tritium Generation

As seen in the figure, tritium is generated through many reactions. Tritium generation rate is calculated using the according models. Tritium generation models can be found in table 1.

2.3 Leakage

The leak rate, S_{leak}, of chemical i, is calculated using the equation 12. Where Cw_i is the source flag of the model, MT is the total mass holdup of the node, L_R is the leakage rate. And x_i is the mass fraction of chemical i in the node.

$$S_{leak,i} = Cw_i \cdot MT \cdot L_R \cdot x_i \quad (12)$$

2.4 Purification

Most of the tritium, in primary loop and secondary loop, is removed in the purification system. The removal rate through purification is expressed as

$$S_{PF,i,j} = F_{PF,He} \cdot \eta_i \cdot x_i. \quad (13)$$

Where $F_{PF,He}$ is the flow rate to the purification system and η_i is the fractional purification efficiency of chemical i in the purification system.

2.5 Permeation

Permeation is the core tritium transport mechanism in most of the reactor systems. Since permeation is the key mechanism in tritium analysis, the SNU tritium code adopts a detailed permeation model unlike other existing tritium analysis codes, which use a simple correlation based on Sievert's law. 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.

The simple correlation based permeation model basically ignores these mechanisms and uses a simply

Table 1 Tritium generation models [6]

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)

built correlation based on Sievert's law. This model is widely used. Most of the existing tritium analysis codes such as THYTAN uses this equation 14 [6].

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

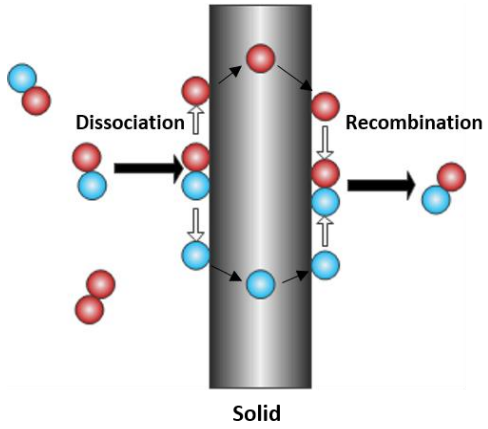

Figure 2 Surface mechanism

For a more precise analysis, the SNU tritium code, BOTANIC, adopts detailed permeation models; equilibrium permeation model and non-equilibrium permeation model. The difference between the equilibrium and non-equilibrium model depends on the surface mechanism assumption. As mentioned, atomic movement on the surface of the solid is governed by the dissociation and recombination processes. Equilibrium assumption assumes that the rate of dissociation and recombination are identical. In other words, there is no atomic flux to the solid surface. A simple solution law is used as seen in equation 15. And in non-equlibrium situation, this assumption does not apply. Conservation equation considering the dissociation rate, recombination rate and the diffusion rate from the surface to the inner solid structure is used to calculate the surface concentration. The non-equilibrium surface model is shown below in equation 16 [7].

$$C_i^{\,2} = K_i \cdot P_m \quad (15)$$

$$\sum_j a_{m_s}\left(K_{d_m} P_m - \sum_{i,j} K_{r_m} C_i^{\,2}\right) + D\nabla C_i = 0 \quad (16)$$

For the permeation movement across the solid structure an atomic conservation equation is used. The conservation equation considers a diffusion term, calculated by Fick's second law, and the decay, trapping and release terms as shown in equation 17 [7]. This model applies to both equilibrium and non-equilibrium situations. The difference between the equilibrium and non-equilibrium assumptions only lies in the surface situation. Furthermore, the detailed permeation model can be solved in 2-dimensional model. To allow a more accurate calculation, a distributed model is solved in the heat exchanger component model.

$$\frac{dC_i}{dt} = -\nabla^2 C_i + S_i - \left\{\frac{\alpha_{t_i} C_{tk}^{*}}{N} C_i - (\alpha_{rk} + v_i) C_i^{tk}\right\} \quad (17)$$

3. Behavior of Tritium Analytic Code (BOTANIC)

3.1 Overview

The Behavior of Tritium Analytic Code, BOTANIC, is developed using a chemical process code called gPROMS. BOTANIC is originally developed for tritium analysis in the fission reactor systems. Since it is based on a chemical process code it has the capability of analyzing chemical process, system dynamics and multi-dimensional tritium analysis.

One of the most distinctive characteristics of BOTANIC is the fact that it does not use a diluted

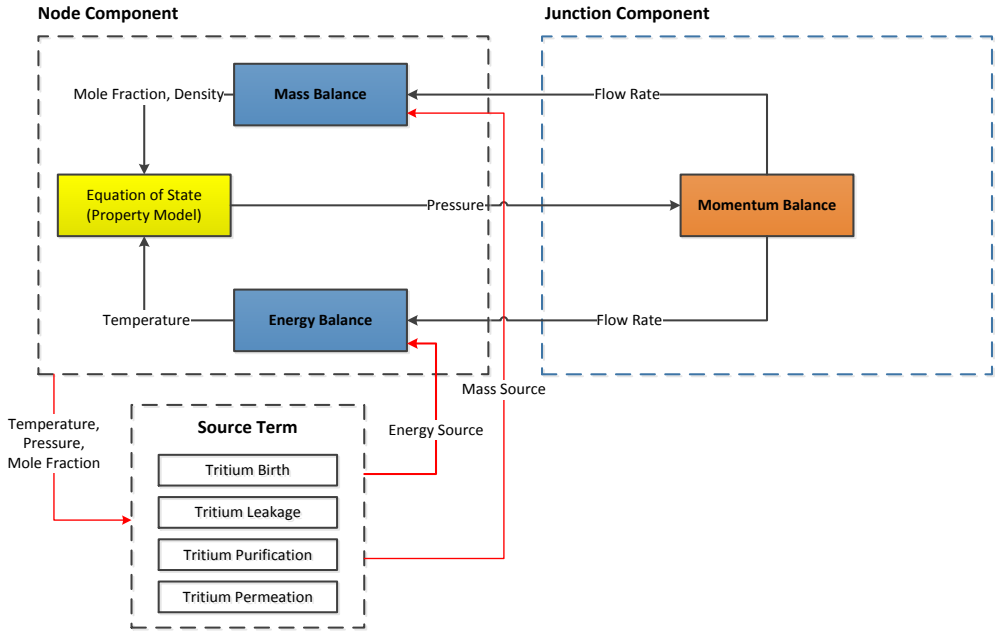

Figure 3 BOTANIC code structure

assumption. Diluted assumption is used in all the existing tritium codes where one assumes that tritium level is extremely low, low enough that any generation or loss of tritium has no effect on the overall physical properties. 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 distributed 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

BOTANIC library is developed using the Process Model Library in gPROMS and is composed of components such as pipe, pump, heat exchanger, etc. Each component is capable of solving mass, energy, or momentum equations depending on its function. Typically node components solve mass and energy conservation equations and junction components solve momentum conservation equation. Associated tritium models are inserted in each component. 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.

4. Verification of BOTANIC

4.1 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 reference data from Peach Bottom core-2, 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 [6], 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 [6]. 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.

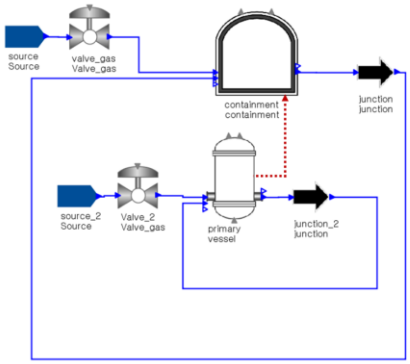

Figure 4 Leakage flowsheeting model

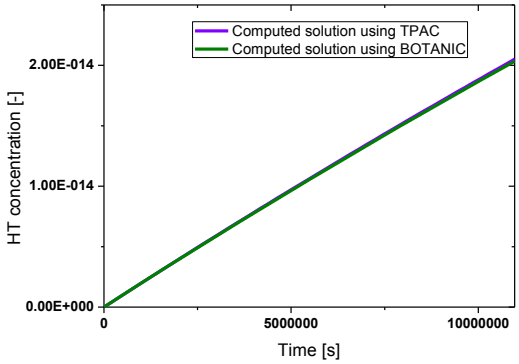

Figure 5 Leakage verification result

4.3 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} [6]. 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.

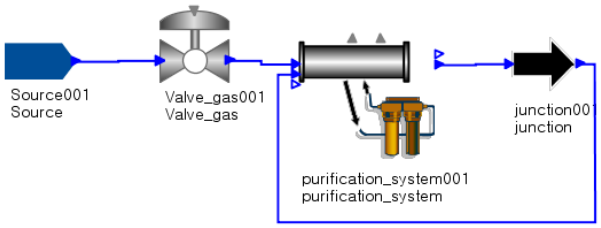

Figure 6 Purification flowsheet modelling

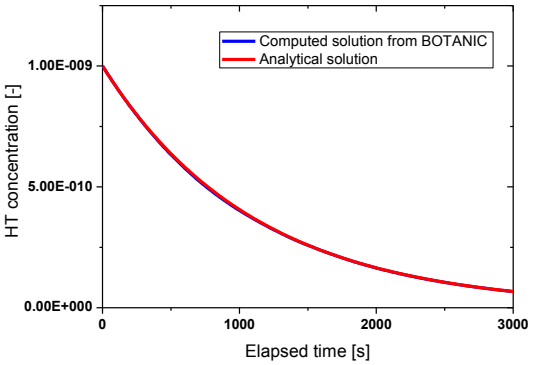

Figure 7 Purification model verification result

4.3 Verification of tritium permeation model

The BOTANIC provides simplified correlation permeation model and the detail permeation model. The Sievert's law based correlation was simply verified using the analytical solution. The permeation rate calculated using BOTANIC was identical to that of the analytical solution.

Table 4 Verification result of permeation model (heat exchanger)

Parameters	Unit	Analytic	BOTANIC
Permeability		1.59×10^{-17}	1.58×10^{-17}
Perm_rate	m3/s	9.48×10^{-13}	9.31×10^{-13}
Perm_rate	kg/s	1.27×10^{-13}	1.25×10^{-13}

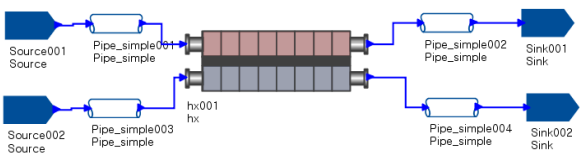

Figure 8 heat exchanger (permeation) verification modelling flowsheet

As the benchmark codes do not provide the distributed permeation model, the distributed permeation model was verified using a multi-physics code called COMSOL. First, in order to verify the 2D permeation model within the solid structure the surface concentration was given with great variance in the y-direction. 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 9. Figure 9 is the concentration of tritium at the center of the wall. As seen in the figure the calculation results seem to be in agreement.

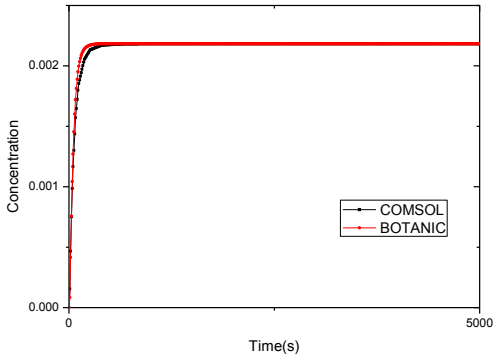

Figure 9 Tritium concentration at the wall center

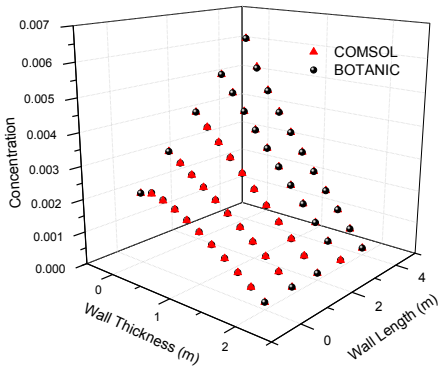

Figure 10 Tritium concentration distribution at steady state

5. Conclusions

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 features including non-diluted assumption, flexible applications and adoption of distributed permeation model. Due to these features, BOTANIC has the capability to analyze a wide range of tritium level systems and has a higher accuracy as it has the capacity to solve distributed models. BOTANIC was successfully developed and verified using analytical solution and the benchmark code calculation result. The results showed very good agreement with the analytical solutions and the calculation results of TPAC and COMSOL. Future work will be focused on the total system verification.

Acknowledgement

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