Design of a Tritium Mitigation and Control System for Fluoride-salt-cooled High-temperature Reactor Systems

THESIS

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By

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Abstract

The Fluoride salt-cooled High-temperature Reactor (FHR) is a Generation-IV molten salt reactor. However, tritium generation from neutron activation of the primary coolant FLiBe (a mixture of molten LiF and BeF₂) is a significant issue in current development of FHRs. Currently a three-loop FHR is under consideration with an intermediate loop for tritium management. As the increased size and therefore the cost of FHRs brought by the additional loop offset the advantages of FHRs, a tritium control system that could be applied to a two-loop FHR is investigated in this study.

In this paper, a tritium control and mitigation system is presented. It consists of three major facilities: the redox control facility, the tritium removal facility and the IHX with tritium permeation barrier. In addition, Al₂O₃ coating as a surface treatment on structural materials is applied as needed to prevent potential tritium leakage. The designs of the facilities are developed and optimized via computational simulation using COMSOL Multiphysics and MATLAB.

The system proposed can achieve three major tasks. The first one is the control of TF existence in the primary coolant, which is highly corrosive to structural materials in the primary loop. The second task is the extraction of tritium from the primary coolant. It would be beneficial if the tritium extracted is collected for further use. The third task is the prevention or minimization of tritium permeation out of the primary loop through intermediate heat exchanger or structural materials.
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Nomenclature

\( A \)  
area if the reactor cross section

\( A_m \)  
mass transfer area

\( A_{ratio} \)  
ratio of pipe ID to the SS316 ball diameter in the packed bed scrubber

\( a \)  
surface area

\( \text{avg} \)  
average neutron energy

\( C_{\text{Be}}^{\text{sat}} \)  
saturation concentration of Be in FLiBe

\( c \)  
concentration

\( c_p \)  
specific heat capacity

\( D \)  
diffusivity

\( D_h \)  
hydraulic diameter

\( d \)  
thickness

\( E \)  
activation energy

\( f \)  
reaction rate

\( \text{Gr} \)  
Grashof number

\( g \)  
gravitational acceleration, 9.8 m/s\(^2\)

\( H \)  
Henry’s constant
\( h \)  
height

ID  
inner diameter

\( J \)  
mass flux

\( K \)  
overall mass transfer coefficient

\( K_p \)  
partition coefficient

\( k \)  
mass transfer coefficient

\( k_{\text{abs}, \text{H}_2\text{Ag}} \)  
chemical reaction rate constant

\( k_t \)  
thermal conductivity

\( k_B \)  
Boltzmann constant

\( k_{\text{BeF}_2} \)  
modified first-order rate constant of chemical reaction

\( k_r \)  
first-order rate constant of chemical reaction

\( L \)  
characteristic length

\( l \)  
length in the axial direction

\( m \)  
mass flow rate

\( m_{\text{HF} \rightarrow \text{H}_2} \)  
chemical reaction rate (HF conversion to H\(_2\))

\( \text{Nu} \)  
Nusselt number

\( n_{\text{tot}} \)  
total amount of chemicals expressed in moles

\( th \)  
thermal neutron energy

\( T \)  
temperature

\( t \)  
time

\( t_w \)  
wall thickness
OD  outer diameter

$P$  permeability

Pe  Peclet number

Pr  Prantl number

$p$  pressure, or partial pressure

$Q$  mass flow rate

$Q_m$  total mass transfer rate

$R$  radius

Re  Reynold’s number

$R_g$  gas constant, 8.314 J/K-mol

$r$  radial direction vector

$r$  axial in radial direction

$S$  solubility

Sc  Schmidt number

Sh  Sherwood number

St  Stanton number

$u$  velocity vector

$V_r$  volume of reaction area

$v$  average velocity

$x_{Be}$  concentration of Be in chemical reaction

$x_r$  concentration of chemicals in reaction

$x_s$  distance from the molten salt inlet

$\frac{\text{total flow length of molten salt}}{x_v}$
$y_{HF}$ concertation of HF in chemical reaction

$z$ axial in $z$ direction

Greek symbol

$\alpha$ thermal diffusion rate

$\beta$ coefficient of expansion

$\varepsilon$ porosity

$\sigma$ neutron absorption cross section

$\rho$ density

$\mu$ kinematic viscosity

$\nu$ viscous diffusion rate

$\psi_c$ tritium generation source term

superscript

$0$ initial state

subscript

$1$ phase 1, or side 1

$2$ phase 2, or side 2

bulk bulk property

$g$ gas phase

$H$ protium
indicating properties at the liquid phase
inlet
indicating properties at the phase interfaces
overall property
outlet
salt property
tritium
unit volume
wall property
1 Introduction

1.1 Fluoride salt cooled High temperature Reactor (FHR)

The Fluoride salt-cooled High-temperature Reactor, which is also referred to as FHR, is a modified design of the Molten Salt Reactor (MSR), a Generation-IV advanced nuclear reactor. In FHRs coated-particle fuel is used, and high-temperature low-pressure molten fluoride salts are used as coolants.

The concept of the FHR originates from the Molten Salt Reactor Experiment (MSRE) which was constructed and investigated in 1960s by Oak Ridge National Laboratory. To date, no FHR has been built and operated yet. However, there have been some designs of the FHR. The most mature design is possibly the Advanced High-Temperature Reactor (AHTR) [1], which is a pre-conceptual design of a central station type FHR. It is used as a prototype for research purposes.

1.1.1 Molten salt coolant

The first usage of fluoride salt in nuclear reactors was in MSRE, where fluoride salt was used in both the liquid fuel and coolant. The salt selected was FLiBe. The reason for MSRE to use FLiBe is the high thermal and radiation stability. Also, because of its low viscosity, it has good flow properties. Currently FLiBe, a eutectic of LiF and BeF₂, is considered as the candidate for the primary coolant. For the secondary coolant, there are
three main candidates: FLiBe, FLiNaK, which is a mixture of LiF, NaF and KF, and KF-ZrF$_4$, which is a mixture of KF and ZrF$_4$.

<table>
<thead>
<tr>
<th>Composition (%) mol</th>
<th>FLiBe</th>
<th>FLiNaK</th>
<th>KF-ZrF$_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiF-BeF$_2$ (67-33)</td>
<td>LiF-NaF-KF (46.5-11.5-42)</td>
<td>KF-ZrF$_4$ (58-42)</td>
<td></td>
</tr>
<tr>
<td>Metatling point (°C)</td>
<td>460</td>
<td>454</td>
<td>390</td>
</tr>
<tr>
<td>Boiling point (°C)</td>
<td>~1400</td>
<td>1570</td>
<td>~1450</td>
</tr>
<tr>
<td>Density $\rho$ (kg/m$^3$)</td>
<td>2413-0.488T $(800-1080 \text{ K})$ [2]</td>
<td>2579.3-0.624T $(940-1170 \text{ K})$</td>
<td>3416-0.887T [3]</td>
</tr>
<tr>
<td>Viscosity $\mu$ (cP)</td>
<td>$0.0116\exp(3755/T)$ $(873-1073 \text{ K})$</td>
<td>$0.04\exp(4170/T)$ $(873-1073 \text{ K})$</td>
<td>$0.0159\exp(3179/T)$ $(873-1073 \text{ K})$</td>
</tr>
<tr>
<td>Heat capacity $c_p$ (J/kg-K) at 700 °C</td>
<td>2414.2</td>
<td>1882.8</td>
<td>1050.2</td>
</tr>
<tr>
<td>Thermal conductivity $k$ (W/m-K) at 700 °C</td>
<td>1.1</td>
<td>0.90 [4]</td>
<td>0.45</td>
</tr>
</tbody>
</table>

Table 1.1 Properties of molten salt coolant candidates [5]
The melting point of FLiBe is much lower than that of LiF or BeF₂, as shown in the phase diagraph Figure 1.1.

Figure 1.1 Binary phase diagram for FLiBe

1.1.2 TRISO particle fuel

TRISO particle fuel used in FHRs is originally developed in the DRAGON project. A TRISO particle has a fuel kernel in the center, and is coated with four layers of three different materials. The diameter of a TRISO particle is around 1 mm. The four
layers, from center to surface, are porous carbon (C), dense pyrolytic carbon (PyC), ceramic SiC and another layer of PyC. Each layer is designed with functions. Porous carbon layer can provide sufficient void volume to accommodate gaseous fission products and fuel swelling. The SiC layer can endure the high gas pressure from the porous carbon layer and retain fission products inside TRISO particles. The inner PyC layer prevents Cl leakage. The outer PyC layer maintains the shape and integrity of the particle. All of the four layers have the function of retaining fission products. Thus, TRISO particle itself is the first barrier of fission products leakage. In operation, the temperature of TRISO particles should not exceed 1600 °C [6].

Figure 1.2 TRISO particle fuel [7]
1.1.3 Features of FHR

The FHR combines the advantages of previous reactor types. Among them the most significant are its passive safety features. The primary coolant has about 700 °C of temperature margin between the operating temperature and the boiling point. The fuel also has a large temperature margin before failure and a negative temperature reactivity coefficient. Thus, the use of TRISO particle fuel and molten fluoride salt coolant enables an FHR to operate near the atmospheric pressure and have a large margin to fuel failure and coolant boiling [8]. Besides, the core power density of an FHR is lower than that of a typical PWR. In cases of accidents, an FHR can keep the core cooled down by natural circulation. Compared to PWRs, FHRs require lower cost on the safety system. The efficient and cost-friendly safety system is also a major reason that FHRs receive high attention.

1.2 Tritium issue in FHRs

Although FHRs have attractive features compared to current commercial Light Water Reactors (LWRs), significant issues still remain. Among them is the tritium control issue. Tritium is generated during the operation of the FHR and is difficult to retain. Since tritium has serious impacts on the environment, proper control and mitigation of tritium is required.

1.2.1 Tritium toxicity

Tritium is a radioactive isotope of the element hydrogen, and is often denoted by $^3\text{H}$ or T. It is a low energy beta emitter. The nucleus of a tritium atom consists of one proton and two neutrons. Naturally occurring tritium is rare on the earth. The main
sources of tritium are interactions between cosmetic rays and the atmosphere and human activities. Tritium has a decay half-life of 12.32 years, while its specific activity is 9650 Ci/g, or $3.57 \times 10^{14}$ Bq/g.

Similar to protium, the lightest hydrogen isotope which is usually denoted by $^1$H or H, tritium has high transport abilities and is difficult to retain. The high permeation coefficient, which is called permeability of tritium in materials enables it to easily leak out of the containers and get into the environment.

Chemical features of tritium are also similar with that of protium. Thus, tritium tends to combine with hydroxyl radicals and oxygen forming tritiated water (HTO or T$_2$O), and with carbon atoms. As a beta emitter, tritium is not hazardous externally, but it can cause serious health problems if inhaled through breath, ingested through food or drink, or absorbed through skin. Thus, regulatory limits of tritium existence in the environment are made to control the potential health risks.

1.2.2 Tritium production and leakage in FHRs

Tritium is generated in the FHR core from neutron activation of the molten fluoride salt coolant. Several main reactions responsible for tritium production in FHRs are summarized as follows, where $\sigma$ represents the absorption cross section (b: barn, the unit of cross section) for the parent nuclide, subscript $th$ refers to thermal neutron properties and $avg$ refers to average neutron properties.

A main source is lithium-6, which is denoted by $^6$Li. In the primary coolant of an FHR, enriched lithium is used, and more than 99.995 wt% of lithium is $^7$Li. However, although the amount of $^6$Li is small, a large amount of tritium can be produced because of
large neutron absorption cross section of $^6\text{Li}$. $^6\text{Li}$ cross section follows $1/\nu$ dependence. It is also estimated that the $^6\text{Li}$ cross section can even reach 5000 b if the neutron energy is low [9]. On the other hand, the reaction between neutron and $^7\text{Li}$ requires that the energy of the neutron is above 0.546 MeV. And for the reaction between $^{19}\text{F}$ and neutron, the neutron energy required is above 9.5 MeV, which is rare in a nuclear reactor. Therefore, $^6\text{Li}$ is still the main concern in tritium production.

Besides, although the reaction between $^9\text{Be}$ and neutron does not generate tritium directly, it produces $^6\text{Li}$, as shown in Equation (1.4). Therefore, even if initial $^6\text{Li}$ is consumed, new $^6\text{Li}$ will be generated through $^9\text{Be}$.

\begin{align}
^6\text{Li} + n &\rightarrow ^4\text{He} + ^3\text{H} + 4.8\text{MeV}; \\
\sigma_{\text{th}} &= 940 \text{ b} \\
^7\text{Li} + n &\rightarrow ^4\text{He} + ^3\text{H} + n'; \\
\sigma_{\text{avg}} &= 20 \text{ mb} \\
^7\text{Li} + n &\rightarrow ^5\text{He} + ^3\text{H}; \\
\sigma_{\text{avg}} &= 20 \text{ mb} \\
^{19}\text{F} + n &\rightarrow ^{17}\text{O} + ^3\text{H}; \\
\sigma_{\text{avg}} &= 30 \mu\text{b} \\
^9\text{Be} + n &\rightarrow ^4\text{He} + ^6\text{Li}; \\
\sigma_{\text{avg}} &= 36 \text{ mb} 
\end{align}

In MSRE, the tritium production rate was 54 Ci/day at the power level of 7.3 MWth [10]. The rate of tritium production in the 2400 MWth AHTR has been
estimated to be 5000 Ci/day at startup and 500 Ci/day during steady state operation. It is comparable to the tritium generation of CANDU reactors. However, the tritium generated in CANDU reactors readily combines with oxygen or hydroxyl radicals and forms T₂O and HTO. These two compounds behave similarly to water and do not penetrate through structural materials. Thus unless leakage of coolant happens, tritium leakage is not considered an issue in CANDU reactors. The tritium production rate in a typical 1000 MWe commercial PWR is 1.9 Ci/day [11]. This is also the tritium release rate of the PWR since no measure is taken in PWRs to prevent tritium leakage.

The majority of tritium generated in FHR cores exists in the charge state T⁺ and it combines with F⁻ to form TF molecules. The ratio of TF and T₂ depends on the chemical redox potential of the primary coolant [12]. As TF is corrosive to structure materials, it must be removed or converted to less corrosive chemicals after its generation in the reactor core as soon as possible [1].

1.2.3 Tritium storage

T₂ extracted from the primary coolant can be collected and stored for future use. For instance, T₂ can be used as fuel in fusion reactors. Recovery of tritium from the purging gas depends on the store method of tritium. According to DOE [13], the three common tritium forms are: gaseous tritium, metal tritides, and tritiated water. Gaseous tritium at ambient pressure is good source for general purpose use, while metal tritide is the preferred form for storage in most cases. In this study, the extracted tritium exists as in helium purging gas. It is preferable in the aspect of operation to use a metal tritide bed for storage.
Sandia in 1970s designed a uranium bed for laboratory use. It was experimentally tested that air ingress has a limited effects in causing tritium leakage from a uranium bed. Titanium has also been investigated for tritium storage. Titanium hydride is stable at room temperature. Additionally, if exposed in air under controlled conditions, titanium hydride can form an oxide layer on the surface and then become stable in air. Palladium tritide is stable at room temperature and the formation requires an overpressure of 6.7 kPa over the tritide bed.

1.2.4 Current research status of tritium control and mitigation

Research has been carried out on the tritium management in the field of fusion reactor, where tritium is used as part of the fuel. It is necessary to recycle tritium as well as deuterium from the exhaust plasma, and therefore the bred tritium can be recovered from the blanket for subsequent use as fuel [14].

Lithium (Li) was investigated as a candidate for tritium getter by several research groups. Talbot [14] proposed a countercurrent liquid extractor for tritium recovery, where the contacting medium is Li-Bi.

Dolan et al. [15] designed a two-stage vacuum disengager for tritium removal from FLiBe based on HYLIFE-II fusion reactor. The optimum size of the FLiBe droplet in this design is 0.4 mm in diameter. A larger droplet size than this results in slower tritium diffusion rate, while a smaller one affects the flow of FLiBe. Also the authors proposed that using a double-wall steam generator in the primary loop, the intermediate loop could possibility be eliminated, since the tritium leakage rate will be sufficiently low.
A double-wall tube-and-shell heat exchanger was proposed in the AHTR design as an intermediate heat exchanger that couples the primary loop and intermediate loop [16]. Yttrium is filled in the gap between the inner and outer tubes as the tritium getter. The challenge for most types of tritium getters is the reaction of the getter material with oxygen on getter surfaces. The oxidation reactions derogate the performance of tritium getters significantly. One method to avoid the surface fouling is to coat the surfaces with palladium (Pd) or platinum (Pt). However, the coating definitely increases the cost and reduces the expected economic benefits.

The counter-current extraction tower is also a widely investigated idea to extract tritium out from the molten salt. Its basic scheme is shown in Figure 1.3.

![Figure 1.3 Schematic of a counter-current flow extraction tower](image-url)
In a counter-current extraction tower, a purge gas such as helium is bubbled from the bottom of the facility, into and through the molten salt, which flows downward. Under the influence of the purging gas bubbles, T₂ dissolved physically in the molten salt will convert to the gas phase and be carried away by the purging gas. Helium is often selected as the purging gas. The reason is that helium has a larger hydrogen diffusivity than most of other common gas species, as will be explained in Section 2.4.3. Fukada et al. [17] developed a model to determine the height of the tower, which is described by Equations (1.5) – (1.7). In this model, \( h \) and \( h_g \) are height equivalents to the theoretical plates of liquid-phase and gas-phase mass transfer, respectively. \( h \) is the height of the counter-current extraction tower. \( \frac{h_g}{h} \) and \( \frac{h_l}{h} \) are dimensionless and called the numbers of transfer unit. \( a_v \) is the total surface area of the bubbles in a unit volume and the diffusion rate of T₂ through the gaseous boundary layer of the He-D₂ mixture is controlled by \( k_g a_v \) (unit: s\(^{-1}\)). \( p_{T_2} \) is the T₂ partial pressure in the gaseous phase. \( \dot{m} \) represents the mass flow rate. \( \mu \) is the kinetic viscosity and \( \rho \) is the density. Subscripts g and l represent the gas phase and liquid phase, respectively. \( R_g \) is the ideal gas constant and \( T \) is the temperature. The model is given as:

\[
h_g = 3.07 \left( \frac{\dot{m}_g}{\dot{m}_l} \right)^{0.32} \left( \frac{\mu_g}{\rho_g D_g} \right)^{2/3} ; \tag{1.5}
\]

\[
h_l = \frac{1}{430} \left( \frac{L}{\mu_l} \right)^{0.22} \left( \frac{\mu_l}{\rho_l D_l} \right)^{0.5} ; \tag{1.6}
\]
Another method that has been frequently proposed is to use a permeation window extraction facility. The basic schematic of a permeation window extraction facility is shown in Figure 1.4.

\[
h = \frac{m_s R_g T}{k_e a_V P_T} \int_{p_{T_2, in}}^{p_{T_2, out}} \frac{dP_{T_2}}{P_{T_2, t} - P_{T_2}}.
\] (1.7)

![Diagram of permeation window extraction facility](image-url)
The total area needed for the permeation window can be calculated using Equation (1.8), where \( k_{l,0} \) is the overall mass transfer coefficient, \( p_2 \), the partial pressure of \( T_2 \) in the downstream He flow, \( c_{\text{in}} \), the \( T_2 \) concentration in the inlet molten salt, and \( c_{\text{out}} \), the \( T_2 \) concentration in the outlet molten salt.

\[
A = \frac{Q}{k_{l,0} \left( c_{\text{in}} - H_{T_2} p_2 \right) - \left( c_{\text{in}} - H_{T_2} p_2 \right)}
\]  

(1.8)

The advantages of the permeation window extraction facility compared to the counter-current extraction tower include:

1. The permeation window extraction facility could be part of the intermediate heat exchanger. This approach potentially reduces the overall cost.
2. The purge gas is not mixed with the molten salt. This eliminates potential complication of two-phase flow.
3. The amount of purging gas that dissolves in the molten salt can be reduced, by properly choosing the permeation wall material. The permeation window material is expected to have selective permeation properties that enable \( T_2 \) to permeate through while allowing only a negligible amount of purging gas to permeate through.

1.3 Research objectives

The primary objective of this research is to design a tritium mitigation and control system for the FHR system added into the primary loop. The tritium control system will mainly achieve two objectives. The first function is to prevent or minimize tritium
permeation into the secondary side or the environment. Therefore, the concentration of tritium in the secondary side and the process heat application side could be significantly reduced compared to without the tritium control system. Overall, the tritium leakage level in the FHRs needs to be comparable with or better than the tritium release rate of 1.9 Ci/day in PWRs. The second function of the tritium control system is to remove tritium from the primary coolant. Tritium generated in the primary coolant cannot be left accumulated even if it does not leak out of the primary loop. The tritium extracted from the primary coolant can be collected as a commodity for further use, such as to be used as the fuel in fusion reactors.

1.4 Thesis outline

This chapter introduces some background of FHR and the tritium issue in advanced nuclear reactors. Chapter 2 will introduce the basis mass transport theories. Chapter 3 introduces the tritium mitigation and control system proposed to achieve the research objective stated in Section 1.3. As the system consists of four main components, Chapter 4 to Chapter 7 will discuss each component separately in detail. At the end, Chapter 8 concludes the paper and give some future outlook of the current work.
2 Tritium Transport Theories

2.1 Diffusion process and laws

Fick’s laws are used to describe the chemical diffusion process. Fick’s first law relates the mass flux $J$ with the concentration gradient $\nabla c$ as:

$$ J = -D \nabla c. \quad (2.1) $$

$D$ is the diffusion coefficient, also referred to as diffusivity. It has a SI unit of m$^2$/s. The diffusivity of species A in species B describes the ability of A transporting in B due to concentration gradient. Diffusivity is dependent on temperature as well as species’ activation energies and can be written as

$$ D = D_0 e^{\frac{E}{k_B T}}, \quad (2.2) $$

where, $D_0$ is the diffusivity at an infinite temperature, which is the maximum diffusivity of the species studied. $E$ is the activation energy with the unit of J/atom. $T$ is the absolute temperature in the unit of K. $k_B$ is the Boltzmann constant.

Fick’s second law, shown in Equation (2.3), predicts the change of concentration with time $\frac{\partial c}{\partial t}$ due to the diffusion. It is actually derived from Fick’s first law together with the mass conservation equation. It is the same as the diffusion equation written as Equation (2.4), assuming $D$ is constant.
In the FHR case, tritium is generated in the reactor core and exists mainly (>90%) in the form of TF [18]. A redox control step to convert TF to T₂ is necessary since TF is highly corrosive to structural materials. The transport of tritium diatomic molecules T₂ follows different mechanisms under different circumstances. Upon formation, T₂ dissolves in the primary coolant FLiBe and then transports with it around the primary loop. In this stage, both molecular diffusion and turbulent diffusion could be involved depending on the flow regime. The transport equation for the T₂ can be expressed as

\[
\frac{\partial c}{\partial t} = D \nabla \cdot (D \nabla c) - \nabla \cdot (c \mathbf{u}) + \psi_c ,
\]

the left hand side of this equation representing the change of concentration of T₂ as time changes. On the right hand side, the first term is the diffusion term and the second is the convection term due to the flow of the primary coolant. \( \psi_c \) is the source term.

Considering circular pipes used in the primary loop of FHR, Equation (2.5) can be rewritten in the cylindrical coordinate system as

\[
\frac{\partial c}{\partial t} = D \left( \frac{\partial^2 c}{\partial r^2} + \frac{1}{r} \frac{\partial c}{\partial r} + \frac{\partial^2 c}{\partial z^2} \right) - \left( u_r \frac{\partial c}{\partial r} + u_z \frac{\partial c}{\partial z} + \left( \frac{u_r}{r} + \frac{\partial u_r}{\partial r} + \frac{\partial u_z}{\partial z} \right) c \right).
\]

Literature [19] suggests that for hydrogen isotopes, their diffusivities in the same material are inversely proportional to the square root of their molecular masses as:
\[
\frac{D_T}{D_H} = \sqrt{\frac{m_H}{m_T}},
\]  

(2.7)

where, \( m \) is the molecular weight of the chemical of interest, and subscripts H and T refer to the isotopes protium and tritium, respectively.

### 2.2 Interphase mass transfer

When diffusion occurs at the interface of two immiscible materials, the interphase diffusion is described by the two-resistance concept, or the two-film theory [20]. The concentration of the solute on both sides of an interface depends on the chemical potentials of the solute in both phases. Thus the concentration of the solute has a discontinuous jump across the interface. The two-resistance concept is shown in Figure 2.1.

Figure 2.1 Two-resistance concept for interphase mass transfer [20]
Partition coefficient or distribution coefficient is the ratio of the concentrations of the solute on both sides of the interface at equilibrium [21]. It is often determined using experimental methods, or predicted using Quantitative Structure-Property Relationship (QSPR) algorithms [22]. As there is not sufficient data on the partition coefficient of hydrogen and molten salt, it is estimated as the ratio of solubility of the two immiscible materials, as:

\[ P = \frac{S_1}{S_2}. \]  

Equation (2.8)

Lang et al. [23] found the correlation applicable in predicting the partition coefficients of the organic solute 1-octanol.

### 2.3 k-type mass transfer coefficient

Analogous to heat transfer, determination of the solute concentration in the bulk of a solvent is usually practical. Thus, to obtain a concentration profile from the bulk solute concentrations, mass transfer coefficients are used as a counterpart of the heat transfer coefficient in heat transfer problems. The mass transfer coefficient \( k \) is defined as:

\[ J = k_1 (c_{1, \text{bulk}} - c_{1, \text{interface}}) = k_2 (c_{2, \text{interface}} - c_{2, \text{bulk}}), \]  

Equation (2.9)

where, \( J \) is the mass flux across the interface between the two phases. Note that \( k \) is not the equivalent to diffusivity \( D \). Subscripts 1 and 2 represent the different phases, and subscript “interface” represents the concentration of \( T_2 \) at the interface in the corresponding phase, while subscript “bulk” is the concentration of \( T_2 \) in the bulk of the corresponding phase.
A local overall mass transfer coefficient $K$ can be defined. For Phase 1, the overall mass transfer coefficient $K_1$ is defined as:

$$K_1 = \frac{J}{c_{1, \text{bulk}} - c_{1, \text{interface}}}$$  \hspace{1cm} (2.10)

where, $c^*$ is the concentration of T2 in Phase 1 at the equilibrium under the current temperature, partial pressure, etc. Similarly, the overall mass transfer coefficient for the other phase $K_2$ can be obtained.

Using $k$-type mass transfer coefficient defined above, mass transfer process resembles heat transfer process. Counterpart heat and mass transfer parameters are listed in Table 2.1 [20]. By obtaining these mass transfer parameters, the method for heat transfer analysis can also be applied to the tritium transport analysis.
### Table 2.1 Mass transfer parameters

<table>
<thead>
<tr>
<th>No.</th>
<th>Mass transfer</th>
<th>Heat transfer</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Reynolds number</td>
<td>Reynolds number</td>
</tr>
<tr>
<td></td>
<td>( Re = \frac{\rho v D_h}{\mu} )</td>
<td>( Re = \frac{\rho v D_h}{\mu} )</td>
</tr>
<tr>
<td>2</td>
<td>Schmidt number</td>
<td>Prandtl number</td>
</tr>
<tr>
<td></td>
<td>( Sc = \frac{\mu}{\rho D_{AB}} )</td>
<td>( Pr = \frac{c_p \mu}{k_i} = \frac{\nu}{\alpha} )</td>
</tr>
<tr>
<td>3</td>
<td>Sherwood number</td>
<td>Nusselt number</td>
</tr>
<tr>
<td></td>
<td>( Sh = \frac{k D_h}{D_{AB}} )</td>
<td>( Nu = \frac{h D}{k_i} )</td>
</tr>
<tr>
<td>4</td>
<td>Peclet number</td>
<td>Peclet number</td>
</tr>
<tr>
<td></td>
<td>( Pe = Re Sc )</td>
<td>( Pe = Re Sc )</td>
</tr>
<tr>
<td>5</td>
<td>Grashof number</td>
<td>Grashof number*</td>
</tr>
<tr>
<td></td>
<td>( Gr = \frac{g L (\Delta \rho)}{\rho \mu^2} )</td>
<td>( Gr = \frac{g D^3 \rho \beta (\Delta T)}{\mu^2} )</td>
</tr>
<tr>
<td>6</td>
<td>Stanton number</td>
<td>Stanton number</td>
</tr>
<tr>
<td></td>
<td>( St = \frac{Sh}{Re Sc} )</td>
<td>( St = \frac{Nu}{Re Pr} )</td>
</tr>
</tbody>
</table>

*: \( \beta \) = coefficient of expansion

### 2.4 Dissolution process and laws

#### 2.4.1 Henry’s law

Henry’s law describes the solubility of a gas in a non-metal liquid. It states that the amount of gas dissolved in the liquid is proportional to the partial pressure of the gas at equilibrium. It is to be noted that Henry’s law only hold for sufficiently dilute solutions.
Henry’s law has several variations of definition. They fall into two basic categories: the ratio of properties of liquid over gas, or the reversed version, i.e., properties of gas over liquid.

The form of Henry’s law used in this study is shown as:

\[ H = \frac{c}{p}, \]  

(2.11)

where, \( c \) is the concentration of the gas dissolved in the liquid at equilibrium state. \( p \) is the partial pressure of the gas. \( H \) is the Henry’s constant. Therefore, from the law equation, the SI unit for \( H \) is mol/m\textsuperscript{3}-Pa. In literature, \( H \) is also sometimes denoted as \( k_H \), and named temperature-dependent constant or simply the solubility of a gas in a liquid.

### 2.4.2 Sievert’s law

Sievert’s law is used to describe the solubility of a diatomic gas into a metal. It predicts that the solubility is proportional to the square root of the partial pressure of the dissolving gas at equilibrium. The reason for the square root relationship is that gas molecules will dissociate into separate atoms when dissolving in metals. The dissociation reaction of H\textsubscript{2} is shown in Equation (2.12). It takes place at the surface of the metal. Therefore, for diatomic gases that have two atoms in a molecule, such as oxygen, hydrogen and nitrogen, the solubility is proportional to the square root of gas partial pressure.

\[ H_2 \rightarrow 2H \]  

(2.12)

In Sievert’s law shown in Equation (2.13), \( c \) is the concentration of the gas in the metal under equilibrium conditions; \( p \) is the partial pressure of the gas; \( S \) is the
equilibrium constant, which is also referred to as the solubility of the gas in the metal. According to Sievert’s law, the unit for solubility of a diatomic gas in a metal is given mol/m$^3$-Pa$^{0.5}$.

\[ S^2 = \frac{c^2}{p} \]  

(2.13)

### 2.4.3 Transport ability of hydrogen isotopes in different materials

The permeation rate of T$_2$ through a wall/coating is influenced by partial pressure on both sides, temperature, the wall/coating material, the thickness of the wall/coating and the surface condition. Permeability is used to describe the ability of a gas to permeate through a certain material, and is the product of diffusivity and solubility, the coefficient to describe diffusion and dissolution.

The permeation rate of tritium through a solid is determined by partial pressure on both sides, temperature, intrinsic properties of the material, the thickness of the solid and the surface condition. Permeability is used to describe the ability of gas permeation through a solid. For gas permeating through a solid, permeability is the product of diffusivity and solubility, the coefficients to describe the effect of diffusion rate and partial pressure, respectively.

\[ P \left[ \text{mol/m-Pa}^{0.5}\text{-s} \right] = D \left[ \text{m}^2/\text{s} \right] \times S \left[ \text{mol/m}^3\text{-Pa} \right] \]  

(2.14)

The permeation rate measured in flux $J$ [mol/m$^2$-s] can be calculated as:

\[ J = DK \left( \sqrt{P_{\text{in}}} - \sqrt{P_{\text{out}}} \right) / d , \]  

(2.15)
where $p_{\text{in}}$ and $p_{\text{out}}$ are partial pressures on the inflow and outflow sides, respectively, and $d$ represents the thickness of the permeating material.

<table>
<thead>
<tr>
<th>Gas</th>
<th>Molten Salt Species</th>
<th>Solubility [mol/m$^3$ - Pa]</th>
<th>Diffusivity [m$^2$/s]</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>T$_2$</td>
<td>FLiBe</td>
<td>$7.9 \times 10^{-2} \exp \left(-\frac{35 \times 10^3}{R_g T} \right)$</td>
<td>$9.3 \times 10^{-7} \exp \left(-\frac{42 \times 10^3}{R_g T} \right)$</td>
<td>[24]</td>
</tr>
<tr>
<td>H$_2$</td>
<td>FLiBe</td>
<td>$1.13 \times 10^{-3}$ (773 K)</td>
<td>$3.17 \times 10^{-3}$ (873 K)</td>
<td>$3.87 \times 10^{-3}$ (973 K)</td>
</tr>
<tr>
<td>D$_2$</td>
<td>FLiBe</td>
<td>$3.1 \times 10^{-4}$ (873 K)</td>
<td>$1.0 \times 10^{-4}$ (973 K)</td>
<td>$8.0 \times 10^{-10}$ (873K)</td>
</tr>
<tr>
<td>H$_2$</td>
<td>FLiNaK</td>
<td>$3.98 \times 10^{-7} \exp \left(\frac{34.4 \times 10^3}{R_g T} \right)$</td>
<td>$8.69 \times 10^{-10} \exp \left(-\frac{50 \times 10^3}{R_g T} \right)$</td>
<td>[27]</td>
</tr>
<tr>
<td>H$_2$</td>
<td>FLiNaK</td>
<td>$7.06 \times 10^{-5} \exp \left(-\frac{54.9 \times 10^3}{R_g T} \right)$</td>
<td>$1.67 \times 10^{-7} \exp \left(-\frac{27.0 \times 10^3}{R_g T} \right)$</td>
<td>[30]</td>
</tr>
</tbody>
</table>

Table 2.2 Solubility of tritium in molten salts studied via experiments

Palladium (Pd) is a good candidate for the permeation window. Pd is widely applied in membrane hydrogen selection processes. The advantages of using Pd are its high hydrogen permeability, good temperature stability, good corrosion resistance, and high hydrogen selectivity. The diffusivity of hydrogen in Pd is listed in Table 2.3 in comparison with that in alloys and nickel. The values of hydrogen permeability through
Pd at the operating temperature obtained from a variety of experiments agree well with each other with only small deviations. Therefore, it was decided to use one of the correlations derived from experiments [28], as:

\[ P = 1.92 \times 10^{-7} \exp \left( -\frac{13810}{8.3144T} \right). \] (2.16)

<table>
<thead>
<tr>
<th>Material</th>
<th>Permeability $[\text{mol/m-s-Pa}^{0.5}]$</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Incoloy-800</td>
<td>$2.4 \times 10^{-10} - 5.5 \times 10^{-10}$</td>
<td>[29] Röhrig, H.D.</td>
</tr>
<tr>
<td>Incoloy-600</td>
<td>$6.6 \times 10^{-10}$</td>
<td>[29] Röhrig, H.D.</td>
</tr>
<tr>
<td>304L</td>
<td>$1.2 \times 10^{-10}$</td>
<td>[29] Röhrig, H.D.</td>
</tr>
<tr>
<td>Hastelloy</td>
<td>$4.2 \times 10^{-10}$</td>
<td>[29] Röhrig, H.D.</td>
</tr>
<tr>
<td>Nickel</td>
<td>$9.3 \times 10^{-10}$</td>
<td>[29] Röhrig, H.D.</td>
</tr>
<tr>
<td>Pd</td>
<td>$2.6 \times 10^{-8}$</td>
<td>[30] Chen, W.</td>
</tr>
<tr>
<td>Pd</td>
<td>$3.65 \times 10^{-8}$</td>
<td>[28] Morreale, B.D.</td>
</tr>
</tbody>
</table>

Table 2.3 Tritium permeability of structural materials

Table 2.4 shows the diffusion coefficients of tritium in several candidate purging gases. Tritium diffusion coefficients in nitrogen, air and carbon dioxide are on the same order of magnitude $\left(10^{-4} \text{ m}^2/\text{s}\right)$, while that in helium is one order of magnitude higher $\left(10^{-3} \text{ m}^2/\text{s}\right)$. However, the tritium diffusion coefficient in the purging gas has a relatively low impact on the overall tritium removal rate. This is because of the low mass transport resistance of the purging gas. Helium would be a promising candidate as the purging gas.
This is because helium has excellent chemical stability under high temperatures, as well as good accessibility. Air could be a candidate of purging gas, as long as the tritium concentration at the outlet of the facility does not reach the explosion limit and the oxygen does not cause corrosion concerns. In order to minimize heat loss to the purging gas, the purging gas would be heated up to the molten salt temperature before being injected into the system. In the computational simulations performed in this study, both helium and air are used as the purging gas.

<table>
<thead>
<tr>
<th>Gas Species</th>
<th>Correlation ((D \text{ in } [m^2/s], p \text{ in } [atm], T \text{ in } [K]))</th>
<th>Diffusivity at 977 K ([m^2/s])</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>(D = 1.032 \times 10^{-8} T^{1.74}) (-)</td>
<td>(1.64 \times 10^{-3})</td>
<td>[31]</td>
</tr>
<tr>
<td></td>
<td>(D = \frac{1.539 \times 10^{-6} T^{1.548}}{\ln \left( \frac{T}{3.16 \times 10^{-7}} \right)^2 \exp \left( \frac{1067}{T^2} - \frac{2.8}{T} \right) p}) (-)</td>
<td>(6.08 \times 10^{-4})</td>
<td>[32]</td>
</tr>
<tr>
<td>N₂</td>
<td>(D = 3.64 \times 10^{-9} T^{1.75} p) (-)</td>
<td>(5.639 \times 10^{-4})</td>
<td>[32]</td>
</tr>
<tr>
<td>Air</td>
<td>(D = 3.64 \times 10^{-9} T^{1.75} p) (-)</td>
<td>(5.639 \times 10^{-4})</td>
<td>[33]</td>
</tr>
<tr>
<td>CO₂</td>
<td>(D = 3.14 \times 10^{-5} T^{1.75} p) (-)</td>
<td>(5.3 \times 10^{-4})</td>
<td>[33]</td>
</tr>
</tbody>
</table>

Table 2.4 Tritium diffusivity in gases
3 Tritium Control and Mitigation System

The system consists of a redox control station, a tritium removal facility, and an intermediate heat exchanger with a tritium permeation barrier, as shown in Figure 3.1. Also, the tritium removal facility has an additional loop to collect tritium in the purging gas, which is extracted from the primary coolant.

Figure 3.1 Tritium removal system for FHR primary loop
The functions of each facility/station are listed below:

Redox Control: This station adjusts the amount of redox agent, which in this design is beryllium (Be), in the primary loop. The main function of Be in tritium control and mitigation is to control the amount of TF in the primary loop. The amount of redox agent affects the redox state of the primary coolant FLiBe. It should be moderately reducing, and therefore, tritium generated has little probability of forming TF, or the TF formed could be converted to T₂ in a short time period.

Tritium Removal Facility: In this facility, T₂ is extracted out of the primary coolant. The design of this facility is a cross-flow configuration, with FLiBe flowing in the normal direction of a tube bank and the purging gas flowing through the inside of the tubes. T₂ dissolved in FLiBe diffuses into the purging gas and is taken away to a post processing facility, where T₂ can be collected and stored for future uses, for instance, to be used in fusion reactor as the fuel. The store of tritium has been introduced in Section 1.2.3.

IHX with a Tritium Permeation Barrier: In this design to increase the diffusion driving force, T₂ concentration in the primary coolant is kept at a high level. The intermediate heat exchanger is a good permeation window for T₂ because of its large surface area and high temperature condition. Therefore, measures must be taken to prevent T₂ leaking through the IHX. A tritium permeation barrier can be applied to the wall surfaces of the IHX to achieve this objective. The details of each facility or station are discussed in the following sections.
Metal tritide bed: Purging gas with extracted tritium flows by a metal tritide bed after the tritium removal facility. The tritium carried out by the purging gas will be trapped in the metal, and form metal tritide. The material suggested in this study is titanium. Titanium tritide is stable in air at room temperature. The purging gas, which is suggested to be helium in this study, does not react with the bed material. After the metal tritide bed, tritium is separated from the purging gas.

Purging gas heat recuperator: The primary coolant of an FHR operates at around 700 °C. In the tritium removal facility, along with mass transfer, heat transfer also takes place. Due to large surface area of the facility, the heat transfer is expected to be ineligible. Thus, it is preferable to recover or recuperate the heat from the purging gas.
4 Redox Control

The form of tritium existence upon generation in an FHR core depends on the redox condition of the primary coolant FLiBe. If the primary coolant is in an oxidation condition, the major form of tritium will be TF. While if the coolant is a reduction condition, the major form of tritium will be T$_2$. If no measure is taken to the redox condition of the primary coolant, it is estimated that more than 90% of the tritium generated in core exists in the form of TF. The remainder tritium exists in the form of T$_2$. As TF is corrosive to structure materials, it must be removed or converted to less corrosive chemicals after its generation in the core as soon as possible [Error! Bookmark not defined.]. The method used to maintain TF concentration at a sufficiently low level is redox control.

Redox control in its basis, is a chemical measure taken to adjust the oxidative or reducing condition of the salt. Therefore, besides controlling the concentration of TF, redox control also adjusts the oxidative conditions caused by impurities. Impurities are introduced into the salt due to impurities in the structural materials (for instance, the oxide layer on the surfaces of the structural materials), the original impurities in the salt and exposure to air or moisture. Since the metal in the structural materials tends to be reducing, severe corrosion will occur if the salt is in an oxidative condition. Thus, it is desirable to keep the salt in a slightly reducing condition using the redox control method.
In MSRE, the fuel was dissolved in the primary coolant. Thus, the redox control was achieved by maintaining the U^{4+} and U^{3+} concentration ratio in the primary coolant [34]. The function of U^{4+} is to keep the salt in the primary loop in a slightly reducing condition. Therefore the oxidative impurities and the oxidative fission process were under control. On the other hand, the function of U^{3+} is to prevent the salt from becoming more reducing than needed. Therefore, the salt would not corrode non-metal structural materials, such as carbon.

In FHR, since fuel is insulated in the TRISO particles and isolated from the primary coolant, substitutes of the U^{4+}/U^{3+} pair are needed. In fusion reactors there has been a significant amount of research done in this area. The redox agent used is Be, which has been investigated in the JUPITER-II experiment [35]. Be reacts with HF when dissolved physically in FLiBe. The chemical reaction function is

\[
\text{Be} + 2\text{HF} \rightarrow \text{BeF}_2 + \text{T} + \text{HF}
\]

(4.1)

Experiments on redox control have been performed by Fukada et al. [35][36] and Simpson et al. [36]. In some of the experiments H_2 was used as the surrogate of T_2. The advantages of using Be as the redox control material include:

1. The reaction rate of this redox reaction is sufficiently rapid to achieve a targeted low concentration of TF. The modified first-order rate constant is

\[
k_{\text{BeF}_2} = 1.5 \times 10^6 \text{ mol/m}^3\text{-s}.
\]

\[
m_{\text{HF}\rightarrow\text{H}_2} = k_{\text{BeF}_2} x_{\text{Be}} y_{\text{HF}}
\]

(4.2)
Equation (4.2) shows the calculation method of conversion rate of HF to $\text{H}_2$.

In this equation $x_{\text{Be}}$ and $y_{\text{HF}}$ denote the concentration of Be and HF in FLiBe, respectively. In chemistry, a first-order reaction means that the reaction rate depends linearly on the concentration of only one reactant. The rate at which a reactant is consumed in a first-order process is proportional to its concentration. Use $c$ to represent the concentration of a reactant, then the reaction rate can be expressed as

$$\text{Rate} = k_r x_r$$  \hspace{1cm} (4.3)

2. The dissolution rate of Be in FLiBe is also sufficiently rapid to ensure the sustain of the reaction rate. The rate constant is $k_{\text{abs}} = 8.9 \times 10^{-8}$ (Be/FLiBe)/s according to the experiments done by Simpson et al. [36].

3. The reaction product BeF$_2$ originally exists in FLiBe. Therefore, the redox control process does not induce impurities.

In this research, the kinetic model developed by Simpson et al. [37] is adopted, as shown in Equations (4.4)-(4.8):

$$\frac{dy_{\text{HF}}}{dl} = \frac{k x_{\text{Be}} y_{\text{HF}} A}{\dot{n}_{\text{tot}}},$$  \hspace{1cm} (4.4)

which describes the distribution of HF concentration along the reaction area. In this equation $x_{\text{Be}}$ and $y_{\text{HF}}$ represent the concentrations of Be and HF. $A$ is the area of the reactor cross section and $l$ is the length in the axial direction. $\dot{n}_{\text{tot}}$ is the total amount of chemicals expressed in moles.
\begin{equation}
\frac{df}{dt} = 1 - e^{\frac{\Delta V_r}{n_{\text{sat}} \cdot x_{\text{Be}}}}, \tag{4.5}
\end{equation}

which models the conversion of HF, where $V_r$ is the volume of the reaction area.

\begin{equation}
x_{\text{Be}} = x_{\text{Be}}^0 \frac{\dot{y}_{\text{HF}}^0}{2 n_s} \int_{0}^{t} f dt \tag{4.6}
\end{equation}

In Equation (4.6) the change of the concentration of Be, $x_{\text{Be}}$, with time $t$.

Superscript 0 denotes the initial state values. $n_s$ is the amount of molten salt in the reaction area in moles.

Equations (4.4)-(4.6) can be combined and rewritten as:

\begin{equation}
t = \frac{2 n_s}{\dot{n}_{\text{tot}} y_{\text{HF}}} \left( x_{\text{Be}}^0 - x_{\text{Be}} \right) + \frac{\dot{n}}{kV_r} \ln \left( \frac{1 - e^{\frac{\Delta V_r}{n_{\text{sat}} \cdot x_{\text{Be}}^0}}}{1 + e^{\frac{\Delta V_r}{n_{\text{sat}} \cdot x_{\text{Be}}^0}}} \right) \tag{4.7}
\end{equation}

Also the dissolution rate of Be in FLiBe, $\frac{dx_{\text{Be}}}{dt}$, can be calculated using Equation (4.7), assuming the initial concentration of Be in the reactor is 0. In this equation, $h$ is the total length of the redox control reaction area. $C_{\text{Be}}^{\text{sat}}$ is the saturation concentration of Be in FLiBe. $A_{\text{Be}}$ is the surface area of Be when dissolving in FLiBe. In the calculation and simulation, an estimated value will be used as $A_{\text{Be}}$.

\begin{equation}
\frac{dx_{\text{Be}}}{dt} = \frac{h A_{\text{Be}} C_{\text{Be}}^{\text{sat}}}{n_s} \tag{4.8}
\end{equation}

Equations (4.4)-(4.8) describe a method for the calculation of the redox control reaction rate. Given the tritium generation rate in the core, and using the equations listed above, the amount of Be can be determined.
Beryllium readily dissolves in the primary salt FLiBe, the redox control is achieved by adding Be directly into FLiBe. After a certain time period of operation, Be is partly consumed and the redox control is not as effective, more Be is added to FLiBe.

Be replenishment can be achieved by adding Be power into the primary loop or by the submersion of a Be rod into the primary coolant. Adding Be power enables accurate control of the amount, while it requires an opening on the primary loop. The usage of Be rod, on the other hand, is advantages in the aspect of operation. The rod can be raised or lowered mechanically. However, it is difficult to control the amount of Be added in. Comparing the two methods, the rod submission method might be preferable. The difficulty in controlling the replenishment amount can be made up through careful monitor of the redox condition of the primary loop. While an opening on the primary loop is one additional potential leakage spot.

In AHTR, the tritium generation rate is 5000 Ci/day at startup [1]. It equivalents to 0.52 g/day. The amount of Be needed can be calculated from the chemical reaction Equation (4.1). At startup the Be consumption rate is 0.0867 mol/day, which equivalents to 0.78 g/day. Therefore, 1 kg Be can last 3.5 years at the startup tritium production rate. Considering the tritium generation rate will gradually decrease to the 500 Ci/day level under steady operation state, the Be consumption rate will also decrease.

However, in practical, besides the conversion of TF to T₂, redox control should also include the management of oxidative impurities. The Be consumption rate is affected by a number of factors. While sufficient Be should exit in the primary coolant, too much Be leads to issues as well. Excessive Be in the primary loop reacts with graphite and
causes corrosion. Therefore, the amount of Be should be carefully controlled. Further
details are needed to accurately determine the amount and frequency of Be
replenishment, such as the corrosion characteristics of the structural materials and the
potential exposure to air or moisture.
5 Tritium Removal Facility

5.1 Key features of successful tritium removal

A successful tritium removal facility should have the following three features:

1. Large surface area to volume ratio: with a fixed overall mass transport coefficient, the higher the ratio of surface area to volume, the higher the transport rate. Thus, in order to reduce the total volume of the tritium removal facility, a large surface area to volume ratio is preferable.

2. Mixing of molten salt: the primary coolant FLiBe has a relatively small tritium diffusivity, which provides a large mass transport resistance if the tritium removal rate depends mainly on the pure chemical diffusion. However, in turbulent flow, mass transport depends highly on convection, or the Eddy diffusion coefficient. Therefore the disadvantage of small tritium diffusivity in FLiBe is no longer a significant issue.

3. Large tritium concentration difference between molten salt side and absorbing/removing side: the concentration difference provides the driving force for the mass transport. Therefore, the larger the driving force, the higher the tritium removal rate per unit surface area.
The key features mentioned above are concluded from computational simulation using COMSOL Multiphysics software as well as MATLAB coding. The details will be explained in Sections 5.2 and 5.3.

5.2 COMSOL simulation of the cross-flow tritium removal facility

5.2.1 Hydrogen diffusion through tube wall model

The computational simulation of tritium transport starts with a simple 2-D axisymmetric tube mode. The geometric scheme of the model is shown in Figure 5.1.

![Figure 5.1 The geometry of permeation tube model (length unit: inch)](image-url)
In this simulation, a 2-D axisymmetric geometry is used to represent the 3-D structure. FLiNaK, is selected for molten salt and stainless steel 316 is used as the tube wall material. On the outer side of the tube wall, air is used as the purging gas.

Considering the low tritium concentration and diffusion rate, a long tube was expected. In the simulation, the tube length is 3 inches scaled down from the actual length of 210 inches.

<table>
<thead>
<tr>
<th>Boundary Number</th>
<th>Boundary Conditions Applied</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Axial symmetric boundary</td>
</tr>
<tr>
<td>2</td>
<td>Inflow concentration</td>
</tr>
<tr>
<td>3</td>
<td>Outflow</td>
</tr>
<tr>
<td>4 (domain A)</td>
<td>$(-D_1 \nabla c_1 + c_1 u) \cdot n = M \left( c_2 - K_{p,1} c_1 \right)$</td>
</tr>
<tr>
<td>4 (domain B)</td>
<td>$(-D_2 \nabla c_2) \cdot n = M \left( -c_2 + K_{p,1} c_1 \right)$</td>
</tr>
<tr>
<td>5</td>
<td>No flux</td>
</tr>
<tr>
<td>6</td>
<td>No flux</td>
</tr>
<tr>
<td>7 (domain B)</td>
<td>$(-D_2 \nabla c_2) \cdot n = M \left( -c_2 + K_{p,2} c_1 \right)$</td>
</tr>
<tr>
<td>7 (domain C)</td>
<td>$(-D_3 \nabla c_3 + c_3 u) \cdot n = M \left( c_2 - K_{p,3} c_3 \right)$</td>
</tr>
<tr>
<td>8</td>
<td>Inflow concentration</td>
</tr>
<tr>
<td>9</td>
<td>Outflow</td>
</tr>
<tr>
<td>10</td>
<td>No flux</td>
</tr>
</tbody>
</table>

Table 5.1 Boundary conditions of the computer simulation model
Table 5.1 lists the boundary conditions of the model. At the boundaries of the adjacent domains, the concentration of hydrogen is not continuous. Partition coefficient $K_p$ is used in the simulation models to indicate the ratio of concentration of hydrogen in different materials. A “stiff spring” method [38] is applied to model the discontinuous jump of concentration at the phase interfaces (mainly the interfaces between the molten salt and the stainless steel, as well as the interfaces between the stainless steel and the purging gas). This treatment of the interphase diffusion boundaries is also used in other simulation models that will be discussed below. Take the boundary between the molten salt domain and the stainless steel 316 domain for illustration, the equation applied is

$$\left(-D_1 \nabla c_1 + c_1 u\right) \cdot n = M \left(c_2 - K_p c_1\right)$$

(5.1)

Here $M$ is a chosen constant which is large enough to make the difference of concentration in the brackets small enough to be considered zero. Thus Equation (5.2) would be satisfied.

$$c_2 = K_p c_1$$

(5.2)

Under steady state and in the axial direction, Equation (2.1) can be simplified to Equation (5.3). This equation can be used to verify the correctness of the model.

$$\frac{\partial c}{\partial z} = \frac{\partial u}{\partial z}$$

(5.3)

In Table 5.2, the local hydrogen concentration of two cases under different operating conditions are compared. The velocity of molten salt was 2 mm/s and 5 mm/s respectively. According to Equation (5.3), to achieve the same hydrogen concentration, the distance in the axial direction needed is proportional to the velocity. For example,
\( v = 2 \text{ mm/s} \) the hydrogen concentration at \( z = 0.6 \text{ inch} \) should be the same as that at

\( v = 5 \text{ mm/s} \) and \( z = 1.5 \text{ inch} \). The values in the two columns in Table 5.2 are almost the same, thus correlating with Equation (5.3). Thus the correctness of the model is verified.

<table>
<thead>
<tr>
<th>R [in]</th>
<th>Local Concentration of Hydrogen ([\times 10^{-7} \text{ mol/m}^3])</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( v = 2 \text{ mm/s} \quad z = 0.6 \text{ inch} )</td>
</tr>
<tr>
<td>0</td>
<td>1.799</td>
</tr>
<tr>
<td>0.01</td>
<td>1.798</td>
</tr>
<tr>
<td>0.02</td>
<td>1.795</td>
</tr>
<tr>
<td>0.03</td>
<td>1.789</td>
</tr>
<tr>
<td>0.04</td>
<td>1.777</td>
</tr>
<tr>
<td>0.05</td>
<td>1.758</td>
</tr>
<tr>
<td>0.06</td>
<td>1.730</td>
</tr>
<tr>
<td>0.07</td>
<td>1.692</td>
</tr>
<tr>
<td>0.08</td>
<td>1.647</td>
</tr>
<tr>
<td>0.09</td>
<td>1.598</td>
</tr>
<tr>
<td>0.10</td>
<td>1.550</td>
</tr>
</tbody>
</table>

Table 5.2 Effects of molten salt velocity on hydrogen concentration

Figure 5.2 shows the influence of various factors on the concentration of hydrogen in molten salt. When molten salt velocity \( v \) is the only variable, hydrogen concentration decreases with the decreasing \( v \). The magnitude of the decrease in concentration is much larger when the velocity drops from 2 mm/s to 1 mm/s compared
36
to that when the velocity drops from 5 mm/s to 2 mm/s. The velocity influences the time
the molten salt stays in the facility. According to Equation (5.4) longer time (larger $t$)
causes more hydrogen to diffuse out of molten salt per unit tube length.

$$ J \times A \times t = \Delta c $$ (5.4)

Also, the two sets of data points at $v = 1$ mm/s, $z = 1.5$ inch and $v = 2$ mm/s,
$z = 3$ inch in Figure 5.2 almost coincide, verifying the correctness of the model.

Figure 5.2 H$_2$ concentration distribution in radial direction at molten salt domain cross
sections ($r = 0.01$ in)

The radius of the tube containing the flowing molten salt also has a significant
influence on the transport of tritium. As shown in Figure 5.3, in the length studied,
hydrogen concentration decreases only in the periphery, while in the center it stays
constant at the inlet level. As stated above, FLiNaK has a relatively low diffusion coefficient, which means the diffusion rate of hydrogen in FLiNaK has to be taken into account in the process. As the radius of tube containing flowing molten salt increases, the larger the resistance the hydrogen meets in the process of reaching the steel structural wall. In addition, a larger radius causes a decrease in the ratio of surface area to volume, which decreases the hydrogen extraction efficiency.

Figure 5.3 H₂ concentration distribution in radial direction at molten salt domain cross sections ($r = 0.03$ in)

The thickness of the structural wall is another factor which influences the extraction rate of hydrogen. Figure 5.4 shows that a thinner wall has a lower local
hydrogen concentration. It can be easily proved that the thinner the steel structure is, the smaller resistance hydrogen experiences when permeating through the wall. Lower diffusion rate in steel causes a relatively higher concentration in molten salt in the case of 0.5 inch thick steel structure, as shown in Figure 5.2.

![Concentration at cross section (wall)](image)

**Figure 5.4 Distribution of concentration of H\(_2\) in the steel structural wall**

In Figure 5.5, it is shown that averaged hydrogen concentration decreases as the molten salt flows forward. The values are listed in Table 5.3. Comparing the simulation results to the tritium removal goal in Table 5.4, the 210-inch tube obviously is not long enough for the permeation window. Considering the diameter as small as 0.02 inch, and the molten salt velocity as low as 1 mm/s, which is almost creeping flow, the simple tube bundle structure is not a practical design for the tritium removal facility.
Figure 5.5 Distribution of averaged H₂ concentration in molten salt flow direction

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Group 1</th>
<th>Group 2</th>
<th>Group 3</th>
<th>Group 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Molten Salt Domain Radius</td>
<td>in</td>
<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
<td>0.03</td>
</tr>
<tr>
<td>Molten Salt Velocity</td>
<td>mm/s</td>
<td>1</td>
<td>2</td>
<td>5</td>
<td>2</td>
</tr>
<tr>
<td>Steel Structure Thickness</td>
<td>in</td>
<td>0.2025</td>
<td>0.2025</td>
<td>0.2025</td>
<td>0.2025</td>
</tr>
<tr>
<td>Flow Length</td>
<td>in</td>
<td>210</td>
<td>210</td>
<td>210</td>
<td>210</td>
</tr>
<tr>
<td>Tritium Removal Percentage</td>
<td>%</td>
<td>33.88</td>
<td>20.01</td>
<td>10.09</td>
<td>5.98</td>
</tr>
</tbody>
</table>

Table 5.3 Comparison of tritium removal efficiency

<table>
<thead>
<tr>
<th>Reactor type</th>
<th>Power</th>
<th>Production Rate [Ci/day]</th>
<th>Tritium Removal Percentage [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>PWR</td>
<td>1000 MWe</td>
<td>1.9</td>
<td>–</td>
</tr>
<tr>
<td>FHR</td>
<td>2400 MWth</td>
<td>500</td>
<td>99.6</td>
</tr>
<tr>
<td></td>
<td>900 MWth</td>
<td>32.8</td>
<td>94.2</td>
</tr>
</tbody>
</table>

Table 5.4 The goal of tritium removal facility in real situations
This simulation model indicates that diffusion in the molten salt is the rate limiting process. A large ratio of extraction surface area to molten salt volume is needed to achieve the tritium reduction goal.

### 5.2.2 Gas bubbling sparger

In this model, a gas bubbling sparger design for the tritium removal facility is simulated. In this design, the purging gas is injected into the molten salt as bubbles from the bottom, and the bubbles rise upward due to buoyancy and inertia, extracting \( \text{T}_2 \) molecules, which are dissolved in the molten salt, in the process. The molten salt can be stagnant, flowing upward, or flowing downward. However, to maximize the extraction efficiency, we recommend that the salt and gas bubbles move in a counter-current manner. In this way, similar to a counter-current flow heat exchangers, the tritium concentration difference between the molten salt and gas can be maximized.
A two-dimensional (2-D) implicit model was built to evaluate the efficiency of this design and determine the required bubble dimensions and concentrations. A modified form of the COMSOL model “Packed Bed Reactor” [39]. FLiNaK was used for the molten salt and helium for the purging gas (namely, the rising bubbles) in the simulation. For the diffusing gas, H2 was used instead of T2 because of limited data of T2 transport coefficients in materials.

The facility was modeled as a simple square, as shown in Figure 5.6, with the $L$ axis representing the height of the facility, and the $r$ axis the radius dimension of a single helium bubble. For example, T2 concentration at point A is the average T2 concentration in FLiNaK in the flow direction at $l_A$ from the inlet of the facility. Also, it is the T2 concentration at the surface of a single bubble at location $l_A$. While the T2 concentration at point B is the T2 concentration inside the bubble, and the distance from point B to the surface of the bubble is $r_B$. The reactor model and the bubble model are coupled through the flux of H2 transported between molten fluoride salt and purging gas bubbles. This flux is given as a chemical reaction rate, calculated based on the total surface area of bubbles per unit volume. By equating the H2 flux leaving FLiNaK to that entering a single helium bubble, and taking into account the diffusion of H2 in the bubble, the model ensures the mass balance of H2.

The calculation results show that when helium bubbles are 2 mm in diameter, and occupy 50% or higher of the total volume of the molten salt flow path, the facility can give a tritium removal rate of 99%. However, bubbly flow can only contain no more than 15-20% volume fraction of bubbles before the bubbles start to considerably collide and
coalesce, decreasing the mass transfer surface area. According to the simulation results, a 10%-volume fraction of bubbles of 1 mm in diameter in the molten FLiNaK flowing at 0.2 m/s will need a 200-meter flow length to accomplish the required removal of tritium.

The results of this gas sparger model also indicates that the mass transfer area is significant in tritium removal. A large mass transfer area to volume ratio is preferable.

5.2.3 Stainless steel 316 packed bed scrubber

Considering the practical difficulty of the purging gas method, a modified design was formulated. Stainless steel 316 balls replace the gas bubbles in the molten salt. In this method, the stainless steel 316 balls act as the collector of T₂. The use of metal balls avoids the bubble merging issue, thus maintaining a high mass transfer surface area. When the concentration of T₂ inside a stainless steel 316 ball increases, its ability of absorbing T₂ will decrease accordingly. When the T₂ concentration in the stainless steel 316 ball reaches a limiting concentration, it will be removed from the molten salt. For simplicity in this thesis, we refer to this situation as the “saturation” of the stainless steel 316 ball, though the T₂ concentration in the stainless steel 316 ball has not yet reached its real solubility limit. The stainless steel 316 balls can be stagnant or circulating. As the amount of stainless steel 316 balls needed is large, they will be recycled and cleaned up after reaching saturation.

Graphite spheres are also considered as the pellets. However, since T₂ diffusivity in graphite is nearly 1/5 of that in stainless steel, it is theoretically less effective than stainless steel 316 in this application. In addition, graphite is a moderator. Therefore, the usage of graphite requires careful evaluation of the potential influences on neutron flux.
For the computer simulation, the model used is that shown in Figure 5.6. Only the properties of helium are replaced with those of stainless steel. H₂ diffusion in stainless steel 316 is much slower than that in the helium gas bubbles. Thus a decrease in efficiency is expected.

Figure 5.7 and Figure 5.8 present the concentration distribution of H₂ in the molten salt at different times for two different models. The x axis is the length of the pipe in the flow direction of molten salt.

Figure 5.7 H₂ concentration distribution of model #3 (see Table 5.5)
Table 5.5 shows the summary of simulation results of the stainless steel 316 packed bed design. The AHTR pre-conceptual design [1] is used for evaluation of the tritium removal efficiency of the model. In this simulation, two sets of commercial pipe sizes (outer diameter is 20 inch, Schedule 40 and outer diameter is 36 inch, Schedule 40) are used. Aspect ratio $A_{\text{ratio}}$ is the ratio of pipe inner diameter to the stainless steel 316 ball diameter. $\varepsilon$ is the porosity, defined as the volume fraction that is not taken by the stainless steel 316 balls. The stainless steel 316 balls are assumed to have a slightly looser packing pattern compared to the densest packing ($\varepsilon = 76\%$). Therefore $\varepsilon$ is set at 40%. Thus the only dimension unfixed is the total length of designed tritium extraction pipe. By checking the dimension of proposed facility needed to fulfill the cleanup task of the
molten salt flow rate proposed by AHTR pre-conceptual design (99% of tritium removal), the total length is estimated as listed in the right most column of Table 5.5.
<table>
<thead>
<tr>
<th>Model #</th>
<th>$A_{\text{ratio}}$</th>
<th>$\varepsilon$</th>
<th>Pipe Outside Diameter [in]</th>
<th>Schedule</th>
<th>Pipe Inner Diameter [in]/[m]</th>
<th>$R$ [in]/[m]</th>
<th>$Q$ [m$^3$/s]</th>
<th>$v$ [m/s]</th>
<th>Re (for FLiNaK/FLiBe) $[10^4]$</th>
<th>$Q$ by AHTR design [m$^3$/s]</th>
<th>Total Length of Pipe Needed [m]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>50</td>
<td>0.4</td>
<td>20</td>
<td>40</td>
<td>18.814/0.4779</td>
<td>0.309/0.075</td>
<td>0.5</td>
<td>6.975</td>
<td>7/3.625</td>
<td></td>
<td>2200</td>
</tr>
<tr>
<td>2</td>
<td>50</td>
<td>0.4</td>
<td>20</td>
<td>40</td>
<td>18.814/0.4779</td>
<td>0.309/0.0075</td>
<td>0.125</td>
<td>1.7425</td>
<td>1.75/1.53</td>
<td></td>
<td>8800</td>
</tr>
<tr>
<td>3</td>
<td>79.65</td>
<td>0.4</td>
<td>20</td>
<td>40</td>
<td>18.814/0.4779</td>
<td>0.0984/0.0025</td>
<td>0.125</td>
<td>1.7425</td>
<td>0.6/0.302</td>
<td>5.54</td>
<td>8800</td>
</tr>
<tr>
<td>4</td>
<td>50</td>
<td>0.4</td>
<td>36</td>
<td>40</td>
<td>34.5/0.8763</td>
<td>1.74/0.0442</td>
<td>0.125</td>
<td>0.5175</td>
<td>1.0513/0.5229</td>
<td></td>
<td>8800</td>
</tr>
<tr>
<td>5</td>
<td>79.65</td>
<td>0.4</td>
<td>36</td>
<td>40</td>
<td>34.5/0.8763</td>
<td>0.0984/0.0025</td>
<td>0.125</td>
<td>0.5175</td>
<td>0.0803/0.0985</td>
<td></td>
<td>8800</td>
</tr>
</tbody>
</table>

Table 5.5 Simulation results of the stainless steel 316 packed bed model
From the different models in Table 9, it can be seen that for a cylinder packed bed with a 36 inch (0.91 m) outer diameter and 200 m length, it takes about 3 minutes for 0.0098 inch (2.5 mm) radius stainless steel 316 balls to reach saturation. There are two ways to increase the time before the stainless steel 316 balls saturate. The first is to increase the facility cross section area. Since the volumetric flow rate of the molten salt is constant, the flow velocity will decrease accordingly. Thus it takes the stainless steel 316 balls longer to become saturated. The second one is to increase the length. To do this, the pipes can be bent or curved to reduce the size of the facility.

This model again shows that a large mass transfer surface area to volume ratio is a key issue for the tritium removal facility design problem.

### 5.2.4 Finned plate tritium removal facility design

Previous literature review and calculations have shown the necessity of a high surface area to volume ratio for the design of the tritium removal facility. Since the problem is similar to many heat transfer problems, the methods used to increase surface area in heat exchanger design can be applied. The method used in this design is to add fins in the molten salt flow path.

Figure 5.9 shows the cross section view of two flow passages in the facility. The general design of the facility uses the idea of a plate type compact heat exchanger. The base structure material is stainless steel 316. The molten salt flows in the finned semi-circular pipe, and the other pipe contains the purging gas. The diameters of both flow channels are 2 mm, and the pitch of the channels on the same plate is 3 mm. The plate thickness is set to be 0.8 times of the channel diameter [40], namely, 1.6 mm as shown in
Figure 5.9. For modeling simplicity, the fins in the molten salt channels are assumed to be straight fins made of stainless steel 316. Five fins array radially and uniformly in each channel, with 0.66 mm in height and 0.1 mm in thickness. In this design, the mass transfer area per unit volume in the molten salt reaches 738 m²/m³.

Figure 5.9 Cross section view of finned plate tritium removal facility model (unit cell)

For the computer simulation, FLiNaK and air were used as the fluids for the molten salt and the purging gas, respectively. Similar to the previous designs, the helium or air would be heated before entering into the facility. Air could be a candidate of the purging gas as long as the hydrogen concentration at the outlet of the facility does not reach the explosion limit and the oxygen does not cause corrosion concerns. The mean
velocity of the molten salt in the channels is 0.1 m/s, while that of the purging gas is
0.5 m/s. Both flows are in the laminar flow regime.

The simulation results shows that this design fails to achieve satisfactory tritium
removal efficiency. In Figure 5.10, cut views at different positions of H\textsubscript{2} concentration
show the diffusion process. The numbers below each cross-sectional view represent the
non-dimensional flow length.

\[ x_s = \frac{\text{distance from the molten salt inlet}}{\text{total flow length of molten salt}} \]

(5.5)

As can be seen from the figures, the concentration gradient is mainly in the
molten salt domain. As FLiNaK flows forward, H\textsubscript{2} close to the stainless steel surfaces
diffuses out of FLiNaK quickly, while the H\textsubscript{2} concentration in the bulk stays at a
relatively high level. Since the flow is laminar, the flow lacks mixing. Thus the
concentration gradient in the molten salt prohibits the H\textsubscript{2} from being extracted out.
5.2.5 Wavy plate PCHE type tritium removal facility

Figure 5.11 Cross section view of 2-D wavy plate design (unit cell)
Another design proposed is the wavy plate. Similar to the finned plate design, it uses a plate compact heat exchanger as its original model. Figure 5.11 shows the cross section view of the facility. There are five layers in the unit cell of the facility, from top to bottom, the second and forth layer are the base structural plates made of stainless steel 316. On top and bottom of the two plates are the path for the purging gas, while between the plates the molten salt flows.

The thickness of the stainless steel 316 plates is 1.0 mm, as well as the height of molten salt flow path. For simulation simplicity, the curvature of the plates is expressed by a sine function

$$y = 0.3 \sin \left( \pi x^2 + \frac{\pi}{2} \right)$$  \hspace{1cm} (5.6)

FLiNaK and air are used in the model as the materials for the molten salt and the purging gas, respectively. The molten salt flow rate is 0.1 m/s, and the air flow rate is 0.5 m/s in same direction as the molten salt. Both flows are laminar flow. As to the H\textsubscript{2} diffusion settings, no flux boundary condition was assigned to the top and bottom boundaries, and the stiff-spring boundary condition is assigned at the interfaces of adjacent diffusion domains.

Considering of the efficiency of computer simulation, the length (in x direction) of the model is set to be 100 mm. The inlet H\textsubscript{2} concentration is set at the same level with the average T\textsubscript{2} production rate suggested by the AHTR pre-conceptual design, which is $1.8 \times 10^{-7}$ mol/m\textsuperscript{3} [1]. After it reaches steady state, the outlet concentration at the center line of the molten salt flow is used as the inlet concentration for the next calculation. This
process is repeated five times, as shown in Table 5.6. In this way, the H₂ diffusion behavior in a long geometry can be approximately modeled using shortened geometry.

![Figure 5.12 H₂ concentration distribution](image)

| Unit | Inlet Concentration $\times 10^{-7}$ [mol/m³] | Outlet Concentration $\times 10^{-7}$ [mol/m³] | $|\Delta|$ $\times 10^{-7}$ [mol/m³] |
|------|----------------------------------|----------------------------------|----------------------------------|
| 1    | 1.8                              | 1.71                             | 0.09                             |
| 2    | 1.71                             | 1.625                            | 0.085                            |
| 3    | 1.625                            | 1.54                             | 0.085                            |
| 4    | 1.54                             | 1.46                             | 0.08                             |
| 5    | 1.46                             | 1.385                            | 0.075                            |
| ...  | ...                              | ...                              | ...                              |
| Last | –                                 | 0.018 (99% removal)              | –                                |

Table 5.6 Results of the wavy plate model
Figure 5.12 shows the distribution of H₂ concentration in the model at steady state. The red color represents higher concentration of H₂ and the blue color represents lower H₂ concentration. The basic pattern shown in this Figure 5.12 can be applied to any position along the flow direction. It can be explained in details as H₂ concentration distribution in the stainless steel 316 plate is high near the molten salt, but decreases quickly towards the air side. Maximum H₂ concentration in the molten salt shows a wavy curve in accordance with the shape of the stainless steel 316 plates. In the stainless steel 316 domain, near the surface which counters the molten salt flow, H₂ concentration is obviously higher than that near the surface on the other side.

From the results listed in Table 5.6, a decrease in H₂ concentration can be observed after each calculation. The amount of decrease shows a decreasing trend as the number of calculation increases, because as the flow path becomes longer, the concentration difference between the molten salt and the stainless steel 316 plate will decrease. This decreasing concentration difference gives a lower mass transfer flux. However, 0.04 mol/m³ can be assumed as the lower limit of a decrease during each calculation, until the outlet concentration reaches 0.09 mol/m³, which is the goal of 99% tritium removal. Under this assumption, the total length of flow needed is 4.3 m, which appears to be an acceptable dimension for the tritium removal facility.

This distribution of H₂ concentration leads to the conclusion that mixing the molten salt flow is a method to increase the tritium removal efficiency.
5.3 MATLAB calculation using $k$-type mass transfer coefficient

The total volume of the tritium removal facility is a significant aspect for the evaluation of the design. It should be controlled at a reasonable size. For a fixed molten salt inlet area, if only the active removal regions are counted, the volume is proportional to the active length needed for the desired tritium removal level. Another important characteristic for design evaluation is the pressure drop of the molten salt flow.

From simulations using COMSOL Multiphysics both the active length and the pressure drop can be obtained. On the other hand, the correlations of $k$-type mass transfer coefficients, as shown in Section 0, can also be used to calculate the required active length of tritium removal. Pressure drop of the molten salt flow can be calculated using correlations as well. Therefore, to obtain a better evaluation of the design, the results from COMSOL and correlation calculations are compared. Two sets of MATLAB code have been written to estimate the active length and pressure drop. The details of the three calculation methods are shown below:

1. COMSOL Multiphysics computational simulation: Using COMSOL Multiphysics, the tritium concentration decrease of a unit removal cell (usually the simulation model built) can be obtained. Based on this result, the total number of units required to get the desired tritium removal level (99% in this study) can be identified. The total pressure drop of molten salt flow is calculated by multiplying the molten salt pressure drop of a unit cell by the number of units needed.
2. MATLAB code A: A set of MATLAB code (Code A) calculates the molten salt pressure drop using standard fluid flow correlations. The total length used in this MATLAB model was obtained from the COMSOL Multiphysics unit cell model.

3. MATLAB code B: This set of MATLAB code (Code B) is developed based on the mass transfer correlations of $k$-type mass transfer coefficients. This code calculates the total length of the tritium removal facility with correlations. Then this total length of the facility is feed to Code A to obtain the molten salt pressure drop.

5.4 Simulation results of the cross-flow tritium removal facility

The results are obtained with the geometry of a bare tube bank. With COMSOL Multiphysics, a 3-D model of the cross-flow tritium removal facility unit was built, as shown in Figure 5.14. In the model used in this simulation, only 9 tubes are included. Therefore it is only a unit of the entire facility and the results have to be scaled up for the evaluation of the total facility. Stainless steel 316 plates are placed on both the top and bottom sides of the molten salt channel. Molten salt flows in from Face 1, and flows out from Face 2. For Face 3 and Face 4 on the sides, symmetrical boundary conditions are applied. Air is used as the purging gas and flows upward into the tube bank from the bottom side, and exits from the top. It is assumed no tritium leakage from the edges of the steel plates or tubes. All the tritium that enters in the molten salt exits in molten salt from Face 2, or diffuse into the purging gas through tube walls. The total height of the unit
model is 4 mm. Both stainless steel 316 plates are 0.3 mm in thickness. The total length of molten salt channel is ten times the outer diameter of the tubes.

It should be noted that in this unit model for simulation, the inflow and outflow regions for the molten salt are much longer than an actual unit will have. In addition, the tube bank extends beyond the top and bottom of the molten salt channel to eliminate the influence of boundary conditions from adjacent domains, and to ensure fully developed flow of molten salt across the tube bank. The unit cell used for subsequent calculations includes only those portions which house the tube bank.

FLiBe is used as the primary loop molten salt, air as the purging gas, and SS316 as the structure material in this model. The parameters are listed in Table 5.7. It is also
the dimensions of #18 in Table 5.8. This set of dimensions for a unit cell is used for the current simulations of the tritium removal facility.

<table>
<thead>
<tr>
<th>Model Input Parameter</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tube Inner Diameter</td>
<td>mm</td>
<td>5</td>
</tr>
<tr>
<td>Tube Outer Diameter</td>
<td>mm</td>
<td>6</td>
</tr>
<tr>
<td>Tube Bank Pitch</td>
<td>mm</td>
<td>8</td>
</tr>
<tr>
<td>Tube Wall Thickness</td>
<td>mm</td>
<td>0.5</td>
</tr>
<tr>
<td>Molten Salt Flowing Velocity</td>
<td>m/s</td>
<td>0.05</td>
</tr>
<tr>
<td>Air Flowing Velocity</td>
<td>m/s</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Table 5.7 Input parameters of the tritium removal unit model

From diffusion theory, concentration difference is the driving force of mass transfer. Thus, to make tritium extraction more efficient, an elevated tritium concentration has been proposed. The impact of higher tritium concentration level in the molten salt on the length of tritium removal facility was simulated with the tritium removal unit model shown in Figure 5.14.

As molten salt flows through the unit, tritium is constantly removed from it, lowering the mass transfer driving force, and thus causing a decrease in tritium removal efficiency. Thus tritium concentration in molten salt at the inlet face is set as the log mean concentration difference. Figure 5.15 shows the simulation results. \( c_0 \) is the tritium production rate of a FHR under steady state operation [1]. The value of \( c_0 \) is \( 1.8 \times 10^{-7} \text{ mol/m}^3 \).
Figure 5.14 Length of tritium removal required at elevated tritium concentration in the primary loop

As the tritium concentration in the primary loop firstly starts to increase, the total length of the facility needed decreases very quickly. However, after the initial tritium concentration reaches three times of the production rate $c_0$, the decrease in total length slows down. If the initial tritium concentration is five times of $c_0$, the total length is about 5 m according to the simulation results. In this situation, the dimension of the tritium removal facility is reasonable in terms of construction and operation.
Different combinations of facility size, molten salt flow rate, purging gas flow rate have been simulated. The results are shown in Table 5.8, and are also plotted in Figure 5.16.
<table>
<thead>
<tr>
<th>Model Number #</th>
<th>Pitch [mm]</th>
<th>Tube Outer Diameter [mm]</th>
<th>Tube Inner Diameter [mm]</th>
<th>Tube Wall Thickness [mm]</th>
<th>Molten Salt Flow Velocity [m/s]</th>
<th>Inlet Frontal Area [m²]</th>
<th>Facility Volume [m³]</th>
<th>Pressure Drop [×10² kPa]</th>
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</table>

Table 5.8 Pressure drop of molten salt flowing across a tube bank
Figure 5.15 Distribution of pressure drop and facility volume
5.5 Design of the tritium removal facility

The finalized dimensions of the tritium removal facility are determined based on the simulation results, as well as the practical realization. With the molten salt flowing across the tube bundle, the tube wall thickness is expected to endure the pressure from the...
flow. Therefore, SCH 40 tube wall thickness is used. The tritium removal facility is designed with modular concept in mind, in order to meet different tritium removal rate demands. In FHRs the required tritium removal rate can change with power levels. Figure 5.17 shows the CAD drawing of the modular tritium removal facility with dimensions. The length unit in this drawing is in inches. In this drawing, only a quarter of the whole tube bank is presented in the front right corner, while in the real facility the tube bank occupies the entire space of the facility. Multiple modules can be bolted together in a row, increasing the total active length of tritium removal area, or they can be bolted side by side to a main pipe, splitting the molten salt flow (Figure 5.18).

Figure 5.17 Tritium removal modular facilities connected to a main pipe
The dimensions of the tube bank as well as the molten salt velocity are listed in Table 5.9. Two sets are presented since they are both close to the optimized values. The flow rate of the primary molten salt used in the calculation of tube numbers is from AHTPR [1].

<table>
<thead>
<tr>
<th>Total Mass Flow Rate of Molten Salt</th>
<th>Unit</th>
<th>Dimension Set A (#24 in Table 5.8)</th>
<th>Dimension Set B (#25 in Table 5.8)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tritium Concentration in Inlet Molten Salt</td>
<td>mol/m³</td>
<td>1.8 × 10⁻⁶</td>
<td>1.62 × 10⁻⁶</td>
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<tr>
<td>Tritium Concentration in Outlet Molten Salt</td>
<td>mol/m³</td>
<td>1.8 × 10⁻⁷</td>
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</tr>
<tr>
<td>Objective Tritium Removal Rate</td>
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<td>1.315</td>
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<tr>
<td>Tube ID</td>
<td>in</td>
<td>0.824</td>
<td>1.049</td>
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<td>Tube Wall Thickness</td>
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<td>0.133</td>
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<tr>
<td>Tube Bank Pitch</td>
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<td>1.64</td>
</tr>
<tr>
<td>Ratio of ( \frac{\text{pitch}}{\text{OD}} )</td>
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<td>1.25</td>
</tr>
<tr>
<td>Tube Number</td>
<td>–</td>
<td>49971</td>
<td>41365</td>
</tr>
<tr>
<td>Molten Salt Inlet Frontal Velocity</td>
<td>m/s</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>

Table 5.9 Dimensions of the tube bank
The tritium concentration in the primary loop will be elevated to 10 times of its generation rate, in order to provide a high mass transfer driving force. The extra tritium in the primary loop will not permeate into the secondary loop, since the IHX has tritium permeation barriers. Currently two methods are being considered to elevate tritium concentration to an ideal level:

1. A valve is installed to control the usage of tritium removal facility. At the startup of a FHR, the valve is closed for a certain time to let tritium build up. After it reaches the target level, the valve will be opened to let tritium be extracted out of the loop. A tritium concentration monitor could also be installed to check the tritium concentration in the primary loop. When it drops below the ideal range, the valve will be closed again.

2. Tritium removal facility works periodically, regardless of the tritium concentration in the primary loop. In this method, a valve is also needed to control the flow of primary coolant. But it does not have to respond to tritium concentration continuously.

5.6 Mass transfer and pressure drop analysis

Based on the geometry, dimensions and the molten salt flow conditions, mass transfer analysis has been performed. An estimation of the total volume of the facility is obtained via calculation of the mass transfer rate. Pressure drop analysis has also been performed, since liquid flowing across the tube bank intuitively results in high pressure drop.

The overall mass transfer coefficient was calculated using Equation (5.7) [41].
\[
\left( \frac{1}{k_o H_s} \right)^{\frac{E}{E}} = \left( \frac{1}{k_s H_s} \right)^{\frac{E}{E}} + \left( \frac{t_w}{P_w} \left( \frac{P_{1,\text{in}}^{0.5} - P_2^{0.5}}{P_{1,\text{out}}^{0.5} - P_2^{0.5}} \right) \right)^{\frac{E}{E}}
\]

(5.7)

\( k \) is the mass transfer coefficient, and subscripts \( o, s \) and \( w \) represent the overall property, the molten salt property and the wall property, respectively. \( p \) is the partial pressure of tritium gas. \( t_w \) is the wall thickness. \( P \) is the tritium permeability in materials.

Numeric subscripts 1 and 2 denote the salt side and the purging gas side, while subscripts in and out denote the inlet and outlet of fluid (both molten salt and purging gas), respectively.

Note that in Equation (5.7), the mass transfer coefficient on the purging gas side is not taken into consideration. The reason is that the mass transfer coefficients of tritium in helium are large enough that they don’t contribute to the overall coefficients.

Total mass transfer area needed is obtained using

\[
Q_m = A_m k_o \left( \frac{c_{1,\text{in}} - H_s p_2}{c_{1,\text{out}} - H_s p_2} \right) \ln \left( \frac{c_{1,\text{in}} - H_s p_2}{c_{1,\text{out}} - H_s p_2} \right),
\]

(5.8)

where, \( Q_m \) is the total mass transfer rate with the unit of mol/s. \( H \) is the Henry’s constant, which is related to the solubility of tritium in materials.

In this tritium removal facility design, the molten salt flows on shell side and across tube banks. Since there is no exact correlation in literature to calculate mass transfer coefficients of fluid across tube banks, the correlations for Nusselt number (\( \text{Nu} \)) in heat transfer are used to calculate Sherwood number (\( \text{Sh} \)). In heat transfer,
\[ \text{Nu} = C \times \text{Re}^m \times \text{Pr}^{0.36} \]  

(5.9)

where factors \( C \) and \( m \) are both function of the Reynolds number (\( \text{Re} \)) [42].

Equation (5.9) is modified in mass transfer to Equation (5.10) for Sherwood number calculation.

\[ \text{Sh} = C \times \text{Re}^m \times \text{Pr}^{0.36} \]  

(5.10)

The results of the analysis are listed in Table 5.10. The total flow length needed is between 20 and 30 meters. As to the pressure drop, it is at the magnitude of \( 2 \times 10^5 \) Pa, i.e., around 2 atm. Considering the construction of a nuclear power plant, the size and pressure drop of the tritium removal facility are not unacceptable.

<table>
<thead>
<tr>
<th>Unit</th>
<th>Dimension Set A</th>
<th>Dimension Set B</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{Re} )</td>
<td>( 10^4 )</td>
<td>4.64</td>
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<td>Molten Salt Inlet Area</td>
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</tr>
<tr>
<td>Total Mass Transfer Area</td>
<td>m²</td>
<td>( 9.85 \times 10^3 )</td>
</tr>
<tr>
<td>Molten Salt Flow Length</td>
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<td>Pressure Loss</td>
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Table 5.10 Results of mass transfer and pressure drop analysis
6 Double-wall IHX with Tritium Permeation Barrier

In the primary loop of a FHR, the intermediate heat exchanger is located after the tritium removal facility. Due to the high level of tritium concentration in the primary coolant, a tritium diffusion barrier is needed to prevent large amount of tritium from leaking into the secondary loop. The barrier must be located between the primary loop and the secondary loop, which means that the barrier would also participate in the heat transfer. Thus another criterion is that the added barrier and related structure should have limited degradation on the heat transfer performance of the intermediate heat exchanger.

The geometry of such a heat exchanger with barrier would be a normal tube-and-shell heat exchanger. A single tube unit is shown in Figure 6.1. Primary coolant flows inside Tube 1. Right outside each tube, there is a concentric tube (Tube 2) which sandwiches the tritium permeation barrier between itself and the primary side tube (Tube 1). The secondary coolant flows in the gap between Tube 2 and Tube 3. In Table 6.1 the design parameters are listed. Simulation results show that T2 concentration in Gap 2 is close to zero, which means the tritium permeation reduction factor of the molten salt (flowing slowly in Gap 1) is quite high.
Molten salts are good tritium diffusion barriers due to their low tritium transport coefficient. Therefore, one proposed design is to use FLiNaK, which has a lower tritium diffusion coefficient than FLiBe, as the tritium permeation barrier. FLiNaK is designed to flow at a very low speed, for example 10 percent of the FLiBe flow rate. An intermediate
heat exchanger using molten salt as tritium permeation barrier will have a similar structure as shown in Figure 6.2.

Figure 6.2 Structure of an IHX using molten salt as tritium permeation barrier

Another design proposed is to use Al₃O₃, which is widely studied as promising tritium permeation barrier material, as the sandwiched tritium permeation barrier in the intermediate heat exchanger. The only difference of this design compared to the FLiNaK one above is that FLiNaK is replaced by Al₃O₃. According to literature, a major application of Al₂O₃ is to apply it as a coating on surfaces. However, one of the main issues with Al₂O₃ coating is that it is difficult to produce uniform coating without defects. Defects in the coating would greatly reduce its effectiveness as tritium permeation barrier. In addition, the coating cracks under irradiation and corrosion, which again derogates its permeation reduction performance significantly. However, in this proposed geometry, Al₂O₃ is sandwiched between two tubes. In this way, Al₂O₃ distributes
uniformly around the inner tube. As long as thermal expansion or deformation due to radiation does not exceed the crack limits of Al₂O₃, this “coating” has little defects that would damage the performance of tritium permeation barrier.

Compared to FLiNaK, Al₂O₃ has a higher thermal conductivity. Thermal conductivity of FLiNaK is around 1 W/m-K, while that of Al₂O₃ is around 30 W/m-K. Thus heat transfer performance are better with Al₂O₃ barrier, if both barriers are of the same thickness.

Current concern of the Al₂O₃ design is that the barrier may reach saturation after starting operation for some time. Unlike FLiNaK or other type of molten salt tritium permeation barrier which can flow and get cleaned up, Al₂O₃ barrier are expected to last longer before being replaced. It still remains to be examined whether Al₂O₃ can still act as satisfactory tritium permeation barrier in an intermediate heat exchanger, even when it is saturated with tritium.
7 Permeation Barriers for Structural Materials

In the current design of the tritium control system, the tritium concentration in the primary loop is maintained at a relatively high level, in order to ensure the sufficient mass transfer driving force. Therefore, there is still a necessity to take measures to prevent tritium leakage through structural materials in the primary loop, such as loop pipes. A practical way to do this is to apply a coating on the outside surfaces of the structural materials. The coating will function as a tritium permeation barrier. Other essential properties of the coating include high-temperature endurance, radiation endurance, good adherence to base material and the ability to maintain integrity. Several materials have been proposed and investigated as candidates for tritium permeation barrier coatings to be applied to surfaces of structural materials. Among them, the most promising ones are yttrium, alumina, tungsten and molybdenum. Each material has different properties besides the common low tritium permeability. The characteristics of each material are discussed below, in section 7.1-7.4.

7.1 Yttrium

Yttrium (Y) is proposed as a tritium getter in fusion reactors since it is a strong hydride former. Experiments performed by Jones et al. [43] showed that yttrium tritide has probable stability. The yttrium tritide tested in the experiments were prepared via the
direct reaction between Y and T\textsubscript{2} gas, at about 66.7 kPa and 750-800 °C. There are limited data of hydrogen transport coefficients in Y, as shown in Table 7.1.

<table>
<thead>
<tr>
<th>Temperature Range [K]</th>
<th>Diffusivity [m\textsuperscript{2}/s]</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>773</td>
<td>3.69×10\textsuperscript{-10}</td>
<td>[44]</td>
</tr>
<tr>
<td>873</td>
<td>7×10\textsuperscript{-10}</td>
<td>[45]</td>
</tr>
<tr>
<td>723-1223</td>
<td>3.4×10\textsuperscript{-8} \exp\left(\frac{-4.06×10\textsuperscript{3}}{T}\right)</td>
<td>[46]</td>
</tr>
</tbody>
</table>

Table 7.1 H\textsubscript{2} diffusivity in Y

Yttrium oxide is proposed as a tritium permeation barrier that could be applied to the surfaces of structural materials. In experiments performed by Wu et al [47], it was estimated that the best deuterium permeation reduction factor (PRF) of the yttrium oxide coating is about 412-244 at the temperature of 873-973 K. The base material is SS316L. The yttrium oxide coating was prepared using the method of sublimation at 150 °C and deposition at 600 °C. It was also pointed out that the increase of pinholes and cracks, as well as the formation of iron oxide in the coating would derogated its performance.

7.2 **Alumina (Al\textsubscript{2}O\textsubscript{3})**

Aluminum oxide (Al\textsubscript{2}O\textsubscript{3}), or alumina, is widely considered as a promising candidate for the tritium permeation barrier in both fusion and molten salt reactors. It has a high melting point of 2072 °C or 3762 °F.
Serra et al. [48] measured $\text{H}_2$ transport coefficients in alumina via experiments. The correlations are listed in Table 7.2. The temperature range of this study was 1273-1673 K, and the $\text{H}_2$ driving pressure range was 10-100 kPa.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Correlation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diffusivity</td>
<td>$\text{m}^2/\text{s}$</td>
<td>$9.7 \times 10^{-8} \exp\left(-\frac{79.99 \times 10^3}{R \ T}\right)$</td>
</tr>
<tr>
<td>Solubility</td>
<td>mol/m$^3\cdot$Pa$^{0.5}$</td>
<td>$5.5 \times 10^{-3} \exp\left(-\frac{22.54 \times 10^3}{R \ T}\right)$</td>
</tr>
<tr>
<td>Permeability</td>
<td>mol/m$\cdot$s-Pa$^{0.5}$</td>
<td>$3.3 \times 10^{-5} \exp\left(-\frac{97.42 \times 10^3}{R \ T}\right)$</td>
</tr>
</tbody>
</table>

Table 7.2 $\text{H}_2$ transport coefficients in $\text{Al}_2\text{O}_3$ by Serra et al. [48]

The $\text{Al}_2\text{O}_3$ coating showed different PRFs in different experiments because of the effects of wall thickness as well as molecular structure and surface conditions of the coating, as shown in Table 7.3.
<table>
<thead>
<tr>
<th>Thickness [µm]</th>
<th>~1</th>
<th>0.2-0.5</th>
<th>1.4</th>
<th>0.03-0.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>PRF</td>
<td>~1000</td>
<td>&gt;3000</td>
<td>10000</td>
<td>100-1000</td>
</tr>
</tbody>
</table>

Table 7.3 Hydrogen permeation reduction factor (PRF) of Al₂O₃ in literature

One of the most significant issues with surface coatings, including Al₂O₃ tritium permeation barrier coating, is that during the operation cracks and other minor defects are likely to develop. These minor defects on the coating surfaces significantly deteriorate the performance of coatings. Therefore, the preparation method of the coating is one of the key issues of the tritium permeation barrier. A sol-gel method described in one paper by Wang et al. [53] appear to be promising. In this paper, an Al₂O₃ coating prepared by the sol-gel method on the surface of a 50 µm thick Fe-Cr-Al alloy foil produced by JFE Steel Corporation (Japan) was examined and tested. The chemical composition of the substrate alloy was Fe-20Cr-5.5Al-0.1Mn-0.08La (wt.%). The coating showed good surface integrity, uniformed thickness and also adherence to the substrate material.

Experiments by Wang et al. [53] also concluded that high temperature (above 1100 °C) prepares better performance coatings. The reason is that at low temperature (500-650 °C), Al₂O₃ prepared is in amorphous form or γ-Al₂O₃. As the temperature goes up, Al₂O₃ becomes crystalized. α-Al₂O₃ is formed at a temperature of 1100 °C and above. An α-Al₂O₃ coating is more uniform in thickness, performances better in aspects of adherence to the substrate material, and develops less cracks.
7.3 Tungsten

Tungsten (W) is a metal with atomic number 74. It is also known as wolfram. Among all naturally occurring pure metals, W has the highest melting point, which is 3422 °C or 6192 °F.

Hydrogen transport in W has been studied by several researchers. The diffusivity and solubility is listed in Table 7.4. The correlation of hydrogen diffusivity in W determined by Frauenfelder [56] is generally accepted as the most accurate. The reason is that the temperature range used in the experiment is above the range where trapping may occur [54]. W has traps for hydrogen at 135 kJ/mol and 241 kJ/mol.

However, although W is a good tritium permeation barrier, several issues remain to prevent its practical application. The hydrogen recombination rate in W is above $10^{-1} \text{ m}^4/\text{s-mol-H}_2$. With such high recombination rate, on the boundary the actual concentration of tritium is likely to be zero. In addition, tritium might not be able to diffuse into the bulk W fast enough. The probable situation is tritium that enters W will be released form the surface again before it diffuse into the bulk.
<table>
<thead>
<tr>
<th>W purity</th>
<th>Temperature [K]</th>
<th>Diffusivity [m²/s]</th>
<th>Solubility [mol/m³-Pa⁰.⁵]</th>
<th>Permeability</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>3N</td>
<td>673 – 1473</td>
<td>( D = 6 \times 10^{-4} \exp\left(-\frac{24.7 \times 10^3}{R_g T}\right) )</td>
<td>–</td>
<td>–</td>
<td>[55] Kakharov</td>
</tr>
<tr>
<td>3N5</td>
<td>1100 – 2400</td>
<td>( D = 4.1 \times 10^{-7} \exp\left(-\frac{9 \times 10^3}{R_g T}\right) )</td>
<td>( S = 1.47 \exp\left(-\frac{24 \times 10^3}{R_g T}\right) )</td>
<td>–</td>
<td>[56] Frauenfelder</td>
</tr>
<tr>
<td>3N</td>
<td>1573 – 2873</td>
<td>–</td>
<td>–</td>
<td>( P = 1.74 \times 10^{-8} \exp\left(-\frac{33 \times 10^3}{R_g T}\right) )</td>
<td>[57] Aitken</td>
</tr>
</tbody>
</table>

Table 7.4 Hydrogen (H₂) transport coefficient in tungsten
7.4 Molybdenum

Molybdenum (Mo) is a transition metal with atomic number 42. Mo has the sixth highest melting point, 2623 °C or 4753 °F, among all naturally occurring pure metals.

Hydrogen diffusivity and solubility in Mo vary significantly in previous studies. On the other hand, hydrogen permeability in Mo obtained in different studies shows consistence. The permeability of H$_2$ in Mo has been reviewed and collected by Steward [58] as shown in Table 7.5. The average permeability of H$_2$ in Mo was described by Chandler and Walter [59] as shown in Equation (7.1). The permeability $P$ has the unit of mol/m-s-Pa$^{0.5}$. The temperature range of this correlation is 500-1700 K, and the pressure range is 0.1-300 kPa.

$$ P = 2.3 \times 10^{-7} \exp \left( \frac{-9710}{T} \right) $$ (7.1)

The diffusivity and solubility of H$_2$ in Mo obtained by Tanabe et al. [73] is considered relatively more accurate. The wide temperature and pressure range used in the study confirms the effects of temperature and pressure on the transport coefficients. Also data from this study represent the upper limit of H$_2$ diffusivity and solubility in Mo, while overestimation of permeability is avoided.
<table>
<thead>
<tr>
<th>Permeability</th>
<th>Temperature Range [K]</th>
<th>Pressure Range [kPa]</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$3.59 \times 10^{-7} \exp \left( \frac{10100}{T} \right)$</td>
<td>820-1670</td>
<td>0.1-10</td>
<td>[60]</td>
</tr>
<tr>
<td>$22.7 \times 10^{-7} \exp \left( \frac{12280}{T} \right)$</td>
<td>850-1250</td>
<td>100</td>
<td>[61]</td>
</tr>
<tr>
<td>$1.65 \times 10^{-7} \exp \left( \frac{9150}{T} \right)$</td>
<td>900-1500</td>
<td>100</td>
<td>[62]</td>
</tr>
<tr>
<td>$4 \times 10^{-7} \exp \left( \frac{11000}{T} \right)$</td>
<td>900-1200</td>
<td>100</td>
<td>[63]</td>
</tr>
<tr>
<td>$2.4 \times 10^{-7} \exp \left( \frac{9660}{T} \right)$</td>
<td>520 and 620</td>
<td>85</td>
<td>[64]</td>
</tr>
<tr>
<td>$3.62 \times 10^{-7} \exp \left( \frac{10800}{T} \right)$</td>
<td>1100-2000</td>
<td>0.3 and 30</td>
<td>[56]</td>
</tr>
<tr>
<td>$2.33 \times 10^{-7} \exp \left( \frac{9710}{T} \right)$</td>
<td>500-1700</td>
<td>–</td>
<td>[59]</td>
</tr>
<tr>
<td>$1.4 \times 10^{-7} \exp \left( \frac{9300}{T} \right)$ (Single-crystal Mo)</td>
<td>679-1270</td>
<td>0.1-30</td>
<td>[75]</td>
</tr>
<tr>
<td>$3.6 \times 10^{-7} \exp \left( \frac{9700}{T} \right)$ (Polycrystalline Mo)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$5.3 \times 10^{-7} \exp \left( \frac{8760}{T} \right)$</td>
<td>540-910</td>
<td>0.1-300</td>
<td>[65]</td>
</tr>
<tr>
<td>$3.22 \times 10^{-7} \exp \left( \frac{9980}{T} \right)$</td>
<td>523-730</td>
<td>100</td>
<td>[66]</td>
</tr>
</tbody>
</table>

Table 7.5 Hydrogen permeability in Mo
| Diffusivity  
| [m²/s] | Solubility  
| [mol/m³-Pa⁰.⁵] | Temperature  
| Range [K] | Pressure  
| Range [kPa] | Reference |
| --- | --- | --- | --- | --- |
| $4.8 \times 10^{-7} \exp \left( -\frac{37.7 \times 10^3}{R_g T} \right)$ | – | 1123–2023 | – | [67] |
| – | $0.72 \exp \left( -\frac{52.2 \times 10^3}{R_g T} \right)$ | 1178–1794 | 100 | [68] |
| $3.5 \times 10^{-7} \exp \left( -\frac{58.6 \times 10^3}{R_g T} \right)$ | – | 523–2023 | – | [69] |
| $2 \times 10^{-6} \exp \left( -\frac{61.5 \times 10^3}{R_g T} \right)$ | $0.18 \exp \left( -\frac{28.5 \times 10^3}{R_g T} \right)$ | 623–1773 | – | [70] |
| – | $0.36 \exp \left( -\frac{39.8 \times 10^3}{R_g T} \right)$ | 873–1473 | 100 | [71] |
| $1 \times 10^{-6} \exp \left( -\frac{58.6 \times 10^3}{R_g T} \right)$ | $0.72 \exp \left( -\frac{52.2 \times 10^3}{R_g T} \right)$ | 1173–1773 | – | [72] |
| $4 \times 10^{-8} \exp \left( -\frac{22.3 \times 10^3}{R_g T} \right)$ | $3.3 \exp \left( -\frac{37.5 \times 10^3}{R_g T} \right)$ | 500–1100 | 0.001-100 | [73] |

Table 7.6 Hydrogen diffusivity and solubility in Mo
Table 7.6 continued

<table>
<thead>
<tr>
<th>Diffusivity ([m^2/s])</th>
<th>Solubility ([mol/m^3-Pa^{0.5}])</th>
<th>Temperature range [K]</th>
<th>Pressure range [kPa]</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>(6 \times 10^{-8} \exp\left(-\frac{16.3 \times 10^3}{R_g T}\right))</td>
<td>(\exp\left(-\frac{54.7 \times 10^3}{R_g T}\right))</td>
<td>723–1173</td>
<td>100</td>
<td>[74]</td>
</tr>
<tr>
<td>(2 \times 10^{-4} \exp\left(-\frac{75 \times 10^3}{R_g T}\right))</td>
<td>(7 \times 10^{-4} \exp\left(-\frac{2.9 \times 10^3}{R_g T}\right))</td>
<td>673–1473</td>
<td>0.1-30</td>
<td>[75] (Single-crystal Mo)</td>
</tr>
<tr>
<td>(2 \times 10^{-4} \exp\left(-\frac{75 \times 10^3}{R_g T}\right))</td>
<td>(1.8 \times 10^{-3} \exp\left(-\frac{6.3 \times 10^3}{R_g T}\right))</td>
<td>673–1473</td>
<td>0.1-30</td>
<td>[75] (Polycrystalline Mo)</td>
</tr>
</tbody>
</table>
7.5 Selection of the tritium permeation barrier coating material

Comparing the tritium permeation barrier coating materials, Al$_2$O$_3$ is probably a promising choice. It can achieve a high permeation reduction factor under certain operation conditions and without cracks. Also, the application of Al$_2$O$_3$ is relatively mature in industry. Several processes are available for Al$_2$O$_3$ coating, including chemical vapor deposition, hot dip method and sol-gel method. Sol-gel method was reported to form high quality surface coatings by Wang et al. [53]. However, the performance of Al$_2$O$_3$ coating under reactor conditions is still a concern.
8 Conclusions and Recommendation for Future Work

In this study, a tritium control and mitigation system has been designed for the Fluoride-salt-cooled High-temperature Reactors. With this system, the tritium release rate of an FHR can be reduced to the same or lower level than that of the tritium production rate of a PWR. AHTR is used in the design process as the prototypic reactor design.

The tritium mitigation and control system consists of four main facilities or stations:

1. Redox control station: Beryllium has been selected as the redox agent.
2. Tritium removal facility: This facility has a cross-flow heat exchanger configuration in geometry, and a modular configuration to allow flexible adaptation to different tritium removal rates.
3. IHX with a tritium permeation barrier: A double-wall heat exchanger is designed with a FLiBe layer in the annulus. FLiBe flows slowly in the annulus acting as a tritium permeation barrier.
4. Tritium permeation barrier coating on structural materials: Al₂O₃ is selected as a promising candidate for the tritium permeation barrier. It has a high tritium permeation reduction factor, but the integrity of the coating remains a limiting issue in practical applications.
For the tritium removal facility, which is the most significant part of the system, several design concepts have been proposed and simulated. The design concepts are compared below in Table 8.1.

<table>
<thead>
<tr>
<th>Model</th>
<th>Conclusions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas bubbling sparger</td>
<td>Bubble collision and coalescence lower the mass transfer area to volume ratio.</td>
</tr>
<tr>
<td>Packed bed scrubber</td>
<td>Although a high ratio of mass transfer area to volume ratio is achieved, the capacity of stainless steel pellets to absorb tritium is not good.</td>
</tr>
<tr>
<td>Finned plate PCHE design</td>
<td>The lack of mixing in the flow makes molten salt itself a tritium diffusion barrier.</td>
</tr>
<tr>
<td>Wavy plate PCHE design</td>
<td>Simulation results of this design verify the necessity of the mixing of the molten salt flow.</td>
</tr>
<tr>
<td>Cross flow design</td>
<td>This design is proposed based on the previous cases studied. Simulation of this design shows a high tritium removal rate.</td>
</tr>
</tbody>
</table>

Table 8.1 Comparison of design concepts of the tritium removal facility

Based on these comparisons, the cross-flow design is adopted for the tritium removal facility. The reason is that the cross-flow design meets the essential requirements for mass transfer. It is shown in the following aspects:

1. The cross-flow design creates a large mass transfer surface area to volume ratio.
   This is mainly due to the large number of tubes in the tube bank.
2. Molten salt flowing across the tube bank leads to forced mixing of the flow.
   Without a high velocity, the molten salt flow can achieve a high Reynolds number, and finally a high mass transfer coefficient.
This thesis presents a preliminary design of the proposed tritium mitigation and control system, which needs further optimization and experimental validation. Issues that need further investigation include but are not limited to the total volume of the system, actual performance and efficiency of the tritium removal, beryllium concentration required in the primary coolant and performance of the facilities.

Future work of this study will mainly focus on the following aspects:

1. Validation experiments of the cross-flow tritium removal facility. A small scale cross-flow tritium removal facility will be constructed to test its effectiveness of mass transfer. In the experiments, H₂ will be used as a surrogate of T₂.

2. Economic evaluation of the tritium mitigation and control system.

3. Heat recovery in the purging gas coming out from the tritium removal facility. The primary coolant operates at around 700 °C. Potentially, the purging gas can carry away a considerable amount of heat. It would increase the overall efficiency of the reactor to recuperate this portion of heat.

4. Accurate data of tritium transport coefficients in molten salt and structural materials. Since the transport coefficients influences the mass transfer coefficients, it would be beneficial to have an accurate data base of the coefficients.

5. Improvement of the tritium permeation barrier coating. Current drawbacks of the surface coating technology significantly limit the performance of tritium permeation barrier coatings.
6. Further investigation of redox control of the primary coolant. It is necessary for controlling the amount of TF in the primary loop.
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