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# Mass transport analysis for tritium removal in FHRs

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ABSTRACT

Tritium control and mitigation is one of the most significant issues in Fluoride Salt-cooled High-temperature Reactors (FHRs). To address this issue, a cross-flow tritium removal facility has been proposed to remove molecular tritium, $T_2$, from the primary coolant. To model the performance of the design, a method is developed based on the logarithmic mean difference of the square root of the tritium partial pressures on the upstream side and the purging gas side. A computer code has been developed based on this global analysis method to study the effectiveness of tritium removal in such a cross-flow tritium removal facility. The code is benchmarked against experimental data from a hydrogen separation experiment in the literature and compared with the results from a code based on traditional finite volume method. Evaluation of the cross-flow tritium removal facility designed for FHR systems has also been performed. The results show how the key factors, i.e., the fluid flow rates and tritium inlet concentration affect the performance of the facility.



## 1. Introduction

Fluoride Salt-cooled High-temperature Reactor (FHR) is categorized as a class of Generation IV nuclear fission reactors (Locatelli et al., 2013). FHR uses eutectic salt mixtures as coolants and is, in a general sense, a type of molten salt reactors (Olson et al., 2009). Currently, FLiBe, (66.7 mol% LiF-33.3 mol% $BeF_2$), is generally considered as the FHR primary coolant. In addition, two salts are considered as potential coolants in the intermediate loop: FLiNaK (a mixture of LiF, NaF and KF) and KF-$ZrF_4$.

While FHRs possess attractive features, such as enhanced safety with passive safety systems and high power generation efficiency, they face a potentially significant issue, i.e., tritium generation and management (Forsberg et al., 2017; Qin et al., 2018). Tritium is primarily generated in the reactor core from the neutron activation of the primary coolant, FLiBe. The coolant salt is purified and enriched with $^7$Li to 99.995 wt%. However, due to the large amount of salt needed in the primary loop and the large reaction cross section of $^6$Li with neutrons, the amount of tritium generated from $^6$Li-neutron reaction is not negligible. Additionally, $^9$Be also reacts with neutrons to produce $^6$Li. The main reactions of tritium production and their corresponding cross sections are listed in Eqs. (1)–(3):

$$^6_3\mathrm{Li} + \mathrm{n} \rightarrow {}^4_2\mathrm{He} + {}^3_1\mathrm{H} + 4.8\ \mathrm{MeV};\ \sigma_{th} = 940\ \mathrm{b} \quad (1)$$

$$^7_3\mathrm{Li} + \mathrm{n} \rightarrow {}^4_2\mathrm{He} + {}^3_1\mathrm{H} + \mathrm{n}';\ \sigma_{avg} = 20\ \mathrm{mb} \quad (2)$$

$$^9_4\mathrm{Be} + \mathrm{n} \rightarrow {}^4_2\mathrm{He} + {}^6_3\mathrm{Li};\ \sigma_{avg} = 36\ \mathrm{mb}. \quad (3)$$

The generation rate of tritium in the primary coolant is about 1 Ci/MWth-day, which is substantially higher than the 0.015 Ci/MWth-day of an average LWR (Wu et al., 2016). Tritium generation in CANDU reactors is about 1.75 Ci/MWth-day (Stempien, 2015) and is comparable to that in FHRs. However, the most significant difference between the tritium generated in CANDU reactors and in FHRs is its existence form. In CANDU reactor, the main form of the tritium is tritiated water, either HTO or $T_2O$. In FHRs, with effective redox control to reduce the salt corrosion to the primary loop structures, the majority of the tritium in the primary coolant exists in the form of tritium bimolecular gas, $T_2$ (Fukada and Mitsuishi, 1988a). Similar to $H_2$, $T_2$ is highly permeable through metal surfaces, especially under the FHR high operating temperatures (Watson, 1972).

In addition, tritium is a weak beta emitter and will cause acute or chronic health issues if inhaled or ingested. The gaseous tritium, $T_2$,

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**Nomenclature**

*Variables*

| | |
|---|---|
| $A$ | mass transfer area, [$m^2$] |
| $c$ | concentration of gas, [mol/$m^3$] |
| $c_p$ | heat capacity, [J/kg-K] |
| $D$ | diffusivity of the gas, [$m^2$/s] |
| $D_H$ | diffusivity of $H_2$, [$m^2$/s] |
| $d$ | tube diameter, [m] |
| $d_h$ | hydraulic diameter, [m] |
| $H$ | Henry's constant [mol/$m^3$-Pa] |
| $h$ | mass transfer coefficient based on concentration of the gas, [mol/$m^2$-s-$Pa^{0.5}$] |
| $h_p$ | mass transfer coefficient based on partial pressure of the gas, [mol/$m^2$-s-Pa] |
| $h_o$ | overall mass transfer coefficient, [mol/$m^2$-s-Pa] |
| $h_t$ | heat transfer coefficient, [W/$m^2$-K] |
| $J$ | permeation flux, [mol/$m^2$-s] |
| $K$ | Sievert's constant of gas, [mol/$m^3$-$Pa^{0.5}$] |
| $k$ | thermal conductivity, [W/m-K] |
| $k_w$ | mass transfer conductivity of gas in the metal tube wall, [mol/m-s-Pa] |
| $L$ | tube length, [m] |
| $LMSPD$ | logarithmic mean difference of square roots of partial pressures of permeating gas between the primary and secondary bulk fluids, [$Pa^{0.5}$] |
| $l$ | characteristic length, [m] |
| $n$ | quantity of gas, [mol] |
| $P_w$ | permeability of gas in the wall, [mol/m-s-$Pa^{0.5}$] |
| $p$ | partial pressure of gas, [Pa] |
| $R$ | ideal gas constant, [J/mol-K] |
| $S_H$ | solubility of $H_2$, [mol/$m^3$] |
| $T$ | temperature, [°C] or [K] |
| $V$ | molar volume of gas, [$m^3$/mol] |

*Greeks*

| | |
|---|---|
| $\alpha$ | thermal diffusivity, [$m^2$/s] |
| $\beta$ | thermal expansion coefficient, [1/K] |
| $\sigma$ | cross section, [barn] |
| $\delta$ | wall thickness [m] |
| $\mu$ | dynamic viscosity, [kg/m-s] |
| $\nu$ | momentum diffusivity, [$m^2$/s] |
| $\rho$ | density, [kg/$m^3$] |

*Non-dimensional numbers*

| | |
|---|---|
| Gr | Grashof number |
| Nu | Nusselt number |
| Pe | Peclet number |
| Pr | Prandtl number |
| Sc | Schmidt number |
| Sh | Sherwood number |
| St | Stanton number |

*Subscripts*

| | |
|---|---|
| 1 | primary side |
| 2 | secondary side |
| *avg* | average value |
| *b* | bulk fluid |
| *w* | tube wall surface |
| $N_2$ | in nitrogen |
| $CO_2$ | in carbon dioxide |
| *i* | at the flow inlet |
| *o* | at the flow outlet |
| *th* | thermal neutron properties |

tends to convert to tritiated water (HTO) after entering the environment (Qin et al., 2017). The hazard of HTO is 25,000 times of that of $T_2$. As a potential health hazard, tritium mitigation and control in FHRs are essential in order to avoid exceeding the tritium release limits from the reactor system to the surrounding environment (Forsberg et al., 2017). Tritium needs to be prevented from releasing into the surrounding environment, but it cannot be left to accumulate in the primary loop either. Therefore, it is necessary to adopt a mechanism to extract and collect tritium from the primary loop. This situation can be generalized as gas separation from liquid under high temperature and atmospheric pressure conditions. A number of strategies have been proposed for this purpose. Studies in fusion systems have shown that a permeation window is one of the most effective methods for hydrogen extraction from a coolant system (Watson, 1972; Fukada and Mitsuishi, 1988b). Among the permeation-window-type designs for tritium removal, a tritium control system featuring a cross-flow tritium removal facility has been proposed (Wu et al., 2016). To evaluate the design of such a cross-flow tritium removal facility applied in FHRs, it is necessary to analyze the effectiveness of the designed facility. To support the analysis, a literature survey of tritium transport and, more generally, hydrogen isotope transport has been performed first.

Permeation of hydrogen isotopes through metals has been extensively studied (Fisher, 1999). It is of interest not only in fusion reactors, but also in fields of hydrogen separation and hydrogen storage for clean energy research. Their main focuses have been on experimental measurements and theoretical model development. Gorman and Nardella (1962) experimentally measured the permeation constant of $H_2$ through various metal samples over a temperature range of 350–950 °C. Steward (1983) reviewed experiments of hydrogen isotopes permeating through metals and non-metal materials. Correlations for the permeability of hydrogen isotopes in different materials were developed from the experimental data. Deveau et al. (2013) proposed a micro-kinetic model for hydrogen permeation in dense metal membranes based on a detailed investigation of the hydrogen permeation process. Andrew and Haasz (1992) investigated the effects of surface conditions for hydrogen permeation through different metals, and proposed a model that takes into consideration different flux saturation mechanisms (University of Toronto, 1991).

Apart from experiments, studies of hydrogen isotopes permeating through metals focus on the development of correlations as well as computer codes. Segmentation mass transfer calculation methods divide the geometry into meshes and impose local mass balance in each cell. Fukada and Mitsuishi (1988a) developed a computer code for a tritium permeation window in fusion reactors. The code was applied to tritium transport in laminar flow fluids between two parallel plates across the walls. The calculation results showed that both resistances in the wall and fluids need to be taken into consideration. In another study (Fukada and Mitsuishi, 1988b), they proposed a model to calculate tritium transport through a tube-type metal permeation window. In this model, an overall Sherwood number was defined for calculation of the overall mass transfer coefficient as:

$$Sh_o^b = \frac{1}{\left(\frac{1}{Sh_m}+\frac{G}{2W_1}\right)^b + \left(\frac{G}{2}\right)^b \left\{ \left[\frac{1-C_m^{1-n}}{1-n}\ln\left(\frac{1}{C_m}\right)\right]^a + \left[\frac{1-C_m^{1-i\cdot n}}{W_2(1-i\cdot n)}\ln\left(\frac{1}{C_m}\right)\right]^a \right\}^{\frac{b}{a}}}, \quad (4)$$