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Development of semi-empirical model for tritium permeation under non-uniform temperature distribution at heat exchanger tube wall

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ABSTRACT

Prediction and mitigation of tritium permeation is an important safety issue in high temperature gas-cooled reactors (HTGRs) especially for industrial applications such as hydrogen production and process heat. This study investigated the effect of non-uniform temperature distribution on the tritium permeation rate through the heat exchanger wall and improved the prediction capability of the existing models by reflecting it. To be more explicit, the effective diffusivity (D_m) and the effective temperature (T_m) was newly defined for the heat exchanger wall, and the effective weight (x_{eff}) was derived from one-dimensional diffusion equation. Based on the data collected by numerical methods, an empirical correlation for x_{eff} was developed by a linear regression method and it was validated by comparisons with randomly generated separate numerical solutions. As a result, the new permeation model based on the effective temperature (T_{eff}) showed very good agreement with the numerical results within an error of 1.28% on average while the existing model based on the average wall temperature (T_m) showed large discrepancies exceeding 200% in the maximum error. This study concludes that the newly developed tritium permeation model significantly improves the prediction capability on the tritium permeation rate through the heat exchangers. Which is the main tritium transport path in the high temperature reactor and the integrated industrial process systems.

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1. Introduction

A high temperature gas-cooled reactor (HTGR), one of the Gen IV concepts, is designed to operate at a high temperature and it allows one to use nuclear process heat for many other industrial applications including hydrogen production and sea water desalination (DOE, 2002). In the HTGRs, permeation of tritium is an important issue because the tritium produced during the high-temperature reactor operations can easily permeate through the heat exchanger walls, and it may radioactively contaminate the coupled systems (Ohashi and Sherman, 2007; Brad et al., 2004; Kim et al., 2010a).

Tritium is a radioactive isotope of hydrogen with a half-life of 12.32 years (Jacobs, 1968). Ordinary hydrogen (H) comprises over 99.9% of all naturally occurring hydrogen while deuterium (D) and tritium (T) comprise 0.02% and 10^{-16}%, respectively. Typical forms of tritium in the environment are tritiated hydrogen (HT) and tritiated water (HTO). Since tritium is radioactive, a major concern is the health effect of tritium on human body. According to the literature, tritium does not constitute a significant external radiation hazard because of its low-energy beta decay and no gamma radiation (see Table 1). However, uptake of tritium can cause potential health problems associated with cancer induction from cell damage like other radioactive materials. The most likely form of uptake tritium is tritiated water, which has 10 days of biological half-life, and uptake of gaseous tritium is typically little less than 1.0%. In the U.S., Environmental Protection Agency (EPA) and the Nuclear Regulatory Commission (NRC) currently limit the tritium concentration within 3.7×10^{-3} Bq/cm^3 in air and 37 Bq/cm^3 in water (Ohashi and Sherman, 2007).

In the HTGRs, tritium is mostly produced in the reactor core by fission of fuel (e.g., ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu) and various neutron capture reactions involving ^{6}Li, ^{7}Li, ^{3}He, and ^{10}B present in the core and coolant. Detailed mechanisms and equations of tritium birth in the nuclear reactor can be easily found in and referred to many research articles and reports by Jacobs, 1968; Wichner and Dyer, 1979; Ohashi and Sherman, 2007; Gainey, 2009; Kim et al., 2010a, and others. Once the tritium is born in the core, it can easily escape into the coolant in the primary loop where the most of the tritium is removed by purification system and the rest can possibly escape outside the primary loop through permeation and leakage.

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Table 1
Radioactive Properties of Tritium (Jacobs, 1968).

Isotope	Half life (yr)	Natural abundance	Specific activity	Decay mode	Radiation Energy (MeV)		
					alpha (α)	beta (β)	gamma (γ)
H-3	12	$\ll$1%	9800	Beta (β)	–	0.0057	–

The remaining tritium can penetrate through the intermediate heat exchanger (IHX) tubes and can migrate to the secondary loop. In the secondary loop, the secondary purification system removes most of the tritium from the secondary coolant just as in the primary loop. The remainders can either escapes outside or permeate into the coupled industrial systems through process heat exchangers (PHXs). The general tritium pathways in the integrated HTGR systems are illustrated in Fig. 1.

Previously, tritium distributions and behaviours in the HTGRs were evaluated for various operating reactors which include the Dragon reactor in England (Forsyth, 1974), the Peach Bottom reactor in the U.S. (Wichner and Dyer, 1979), and the Arbeitsgemeinschaft Versuchsreaktor (AVR) in Germany (Steinwarz et al., 1968). Those activities provided good insights into the mechanisms of tritium production, transport and release to the environment. At that time, since the main issue was general release of tritium to the environment, no significant consideration was made for tritium behaviours in the coupled HTGR systems. The tritium issue in the coupled HTGR systems was raised quite recently since the Next Generation Nuclear Plant (NGNP) planned to use the HTGR as a heat source to produce hydrogen and process steam. Those who first tackled this issue were Ohashi and Sherman, 2007 who investigated tritium distribution and accumulation in a Next Generation Nuclear Plant (NGNP) coupled with two hydrogen plants based on a high-temperature-electrolysis (HTE) process and a thermochemical Sufur-Iodide (SI) process. They used a numerical code named THYTAN and provided good results and insights for distribution and reduction of tritium in the system components and hydrogen product. More recent researches were made by Kim et al., 2010a, 2010b. They developed a computer code named TPAC using MATLAB/SIMULINK package. Using this code, they analysed dynamic behaviours of tritium in the HTGR and coupled hydrogen production system and evaluated important parameters that affect the calculation results using a global sensitivity analysis. As a result, the heat exchangers were identified to be the most important components and their permeation parameters were evaluated to be the most sensitive parameters. It concluded that tritium permeation at the heat exchanger should be well and precisely evaluated and limited to an allowable level in order to minimise the economic and safety penalties. In this study, non-uniformity of the temperature across the heat exchanger tube wall was taken into consideration in the exiting tritium permeation model in order to improve the analysis of tritium in the HTGR and coupled systems. The following sections describes the details.

2. Effect of non-uniform temperature on tritium permeation rate

In order to analyse tritium behaviours in the HTGR systems, various computer codes have been developed and used as summarised in Table 2. They include TRITGO developed by Oak Ridge National Laboratory (ORNL), TMAP and TPAC by Idaho National Laboratory (INL), THYTAN by Japan Atomic Energy Agency (JAEA), and TBEC by Korea Advanced Institute of Science and Technology (General Atomics, 2006; Kim et al., 2010a; Longhurst, 2004; Ohashi and Sherman, 2007; Yook et al., 2006). In those tritium analysis codes, the tritium permeation in the heat exchanger is generally predicted by the following Sieverts' law without solving the diffusion equations in order to avoid computational burden in the whole system analysis (Ohashi and Sherman, 2007; Brad et al., 2004; Kim et al., 2010a; General Atomics, 2006).

$$R_{HX} = \frac{A}{l} \cdot K(T_m) \cdot \left(\sqrt{P_{h2,1}} - \sqrt{P_{h2,2}}\right) \quad (1)$$

$$K(T_m) = S(T_m) \cdot D(T_m) \quad (2)$$

where

- R_{HX} = Tritium permeation rate [mole/s].
- A = Heat exchanger surface area [m^2].
- l = Heat exchanger tube thickness [m].
- T_m = Average tube wall temperature [K].
- $P_{h2,1}$ = Tritium partial pressure in channel 1.
- $P_{h2,2}$ = Tritium partial pressure in channel 2.
- $K(T_m)$ = Tritium permeability at T_m [mole $Pa^{-0.5}$/m s].
- $S(T_m)$ = Tritium solubility at T_m [mole $Pa^{-0.5}/m^3$]
- $D(T_m)$ = Tritium diffusivity at T_m [m^2/s].

However, since the temperature distribution across the heat exchanger tube wall is not uniform in the actual reactors, using the average wall temperature (T_m) may cause a large error in

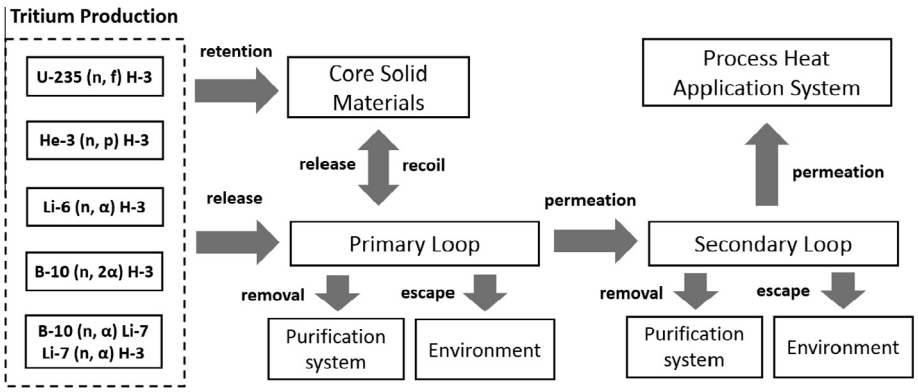

Fig. 1. Tritium production and pathways in the HTGRs.

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