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Tritium permeability measurement in hydrogen-tritium system

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ABSTRACT

Understanding of thermodynamic equilibria of multi-components hydrogen isotopes is required to accurately measure tritium permeability at the expected low tritium partial pressure and non-negligible high hydrogen partial pressure in a fusion blanket system. A gas-driven tritium permeation system that is capable of independently controlling hydrogen and tritium partial pressures was developed at Idaho National Laboratory to accurately measure low partial pressure tritium permeability. The thermodynamic equilibria for hydrogen (H) – tritium (T) permeation through metal are discussed to accurately measure tritium permeability, and the experimental conditions required for evaluating tritium permeability in H-T system are presented.

1. Introduction

Fusion power promises to provide electricity generation with outstanding safety and environmental performance. Safety plays a crucial role in fusion material selection since tritium behavior in materials determines two key safety evaluation source terms: in-vessel inventory source term (i.e. tritium retention) and ex-vessel release term (i.e. tritium permeation), which are used in reactor safety assessments for licensing fusion facilities [1,2]. Tritium permeation through materials at elevated temperature in test blanket system (TBS) is one of major tritium safety issues in ITER and an advanced understanding of tritium behavior in realistic TBS conditions is required to minimize tritium permeation to the port cell. Extensive work on hydrogen and deuterium behavior in fusion materials has been conducted, but only a very small database is available for tritium, the radioactive fuel for future reactors, due to the cost and difficulty associated with handling tritium [3–5]. Hydrogen isotope permeation in the fusion material is mostly investigated with high hydrogen partial pressure system with mono hydrogen isotope ($p_{Q2} > 1000$ Pa where Q = H or D) in the primary side (high hydrogen isotope partial pressure side) and vacuum in the secondary side (low hydrogen isotope partial pressure side) [3–5]. Although these high partial pressure mono-component hydrogen permeation experiments provide variable fundamental databases of hydrogen isotope behavior in high partial pressure (diffusion-limited regime), the surface condition or/and impurities (especially background hydrogen gas) can have a profound impact on tritium behavior in material at low tritium partial pressure [6,7]. The physics of hydrogen (H) – tritium (T) permeation through metal are still poorly understood to confidently design environmentally benign and safe fusion systems at this moment.

Beta detection methods provide excellent detection sensitivities of tritium, enabling us to investigate permeation behavior at low tritium partial pressure, which cannot be achieved using either hydrogen or deuterium. In-line ionization chambers are one of most widely used diagnostics to measure tritium (HT or HTO) in gaseous environments from room air tritium monitoring to tritium detection in permeation experiments [8–10]. Ionization chambers provide superb detection sensitivity (e.g. $\sim 1 \times 10^{-6}$ Ci/m^3 for 1000 cc chambers) and linear response in the wide range of tritium concentration from 0.386 parts per trillion ($\sim 1 \times 10^{-6}$ Ci/m^3 at standard temperature and pressure, STP) to pure tritium ($\sim 2.589 \times 10^{6}$ Ci/m^3 at STP). Special care, however, is needed when the ionization chambers are used for tritium permeation measurements at low partial pressure. When pure helium gas was used as a sweep gas in low tritium partial pressure, the equilibrium was not achieved after 30,000 s following the start of the tritium permeation test of Japanese reduced activation ferritic/martensitic steel (RAFM) F82H at 673 K [11]. The tritium permeation should have been equilibrated after 100's of seconds, but the extra time was required for the tritium molecules to reach surface equilibrium with the dissolved tritium atoms along the inner surface of the tubing connecting the permeating surface to the detector system. Others also observed the effects of tritium dissolved on the wall of the tubing between the permeating surface and detector system, and it is common practice to add a trace amount of hydrogen gas (H_2) in a sweep gas to obtain a faster ionization chamber detection response [12,13]. Additional care is needed when a trace amount of H_2 is used in a sweep gas during a tritium permeation experiment since the existence of H_2 creates multi-hydrogen isotope system in both the primary and secondary sides.

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When the trace amount of H_2 is only added in the secondary side sweep gas, H permeates from the secondary side to the primary side, creating H-T counter-permeation system [14]. In order to accurately measure tritium permeability in the H-T system, one must measure both H_2 partial pressure (p_{H2}) and T_2 partial pressure (p_{T2}) in both the primary and secondary sides. The poor detection sensitivity of hydrogen (> 1 parts per million) often poses a critical challenge in measuring low H_2 partial pressure in the H-T system.

As an example of practical applications, a 1000-ppm (0.1%) hydrogen-helium gas mixture is used as a purge gas at total pressure of 101,325 Pa in the Korean (KO) helium cooled ceramic reflector (HCCR) Test Blanket System (TBS) [15]. The estimated tritium partial pressure in the purge gas stream is less than 10 Pa, and the p_{H2} is higher than the produced HT partial pressure (p_{HT}, tritium is mostly as HT form due to $p_{H2} > > p_{HT} > > p_{T2}$) in the KO TBS purge gas. Improved understanding of tritium permeation behavior in RAFM steel under the H-T system is required to minimize the uncertainty associated with tritium release to the rooms that the TBS pipes pass through (e.g. TBS port cell, Tokamak cooling water systems vault annex) in the Tokamak building for ITER TBS safety. It is important to note that it is challenging to accurately estimate tritium permeability from the H-T system (typically low T_2 partial pressure and high H_2 partial pressure) to negligibly low hydrogen isotope partial pressure due to the decrease of H_2 partial pressure in the H-T co-permeation. Here, we intentionally maintained the identical H_2 partial pressures in both permeation sides, and developed an experimental setup and theory to accurately measure tritium permeability in the H-T system.

In this paper, we discuss the development of a tritium permeation system for low tritium partial pressure in Section 2, the theory of tritium permeability measurement in the H-T system in Section 3, and the required experimental condition for accurately measuring T permeability in Section 4.

2. Experiment

2.1. Experimental apparatus

The Tritium Gas Absorption Permeation (TGAP) system is a vertically standing permeation system that was designed to study tritium absorption and permeation in both solid disc (such as RAFM and tungsten) and liquid materials (such as lead lithium eutectic) and is contained within a ventilated enclosure to minimize worker's tritium uptake [16,18]. The permeation experiment test section consists of two solid gold (Au) wire-rings that are used to seal a disc sample between two custom-made 54.0 mm OD 316 stainless steel flanges as shown in Fig. 1. The test section is installed inside quartz tubing to minimize

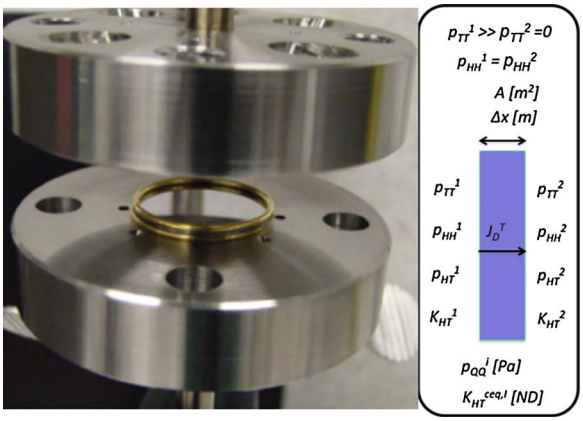

Fig. 1. (Left) Photo of TGAP test section. Two solid gold (Au) wire-rings to seal disc a sample between two custom-made "54.0 mm" OD stainless steel flanges. (Right) Schematic of tritium permeation in H-T system.

tritium permeation to the ventilated enclosure. Gold has the second lowest tritium permeability in metal next to tungsten, and solid Au wire rings (e.g. wire diameter of 1.0 mm) minimize tritium permeation through the O-rings to the quartz tubing [3–5]. A series of COMSOL tritium modeling was performed to understand tritium permeation loss through the test section to the quartz tubing [17,18]. The COMSOL modeling confirmed that the tritium permeation loss to the quartz tubing was negligible for the typical experimental conditions [17,18]. Three test sections were fabricated for testing three different sample sizes (OD of 6.0, 10.0, and 20.0 mm). The sample thickness is typically between 0.5 and 2.0 mm.

Four thermocouples (TC) are inserted (two TC from top and two TC from bottom) to measure the sample temperature at four different locations. The four TCs (TC0, TC1, TC2, and TC3) readings typically agree within 5 K, and one TC, (TC0) is used to control the tube furnace power and the sample temperature by means of a National Instrument (NI) LabVIEW data acquisition and control system. The TC0 is typically maintains the desired sample temperature within ± 2 K for the duration of the tritium permeation experiment. Maximum achievable temperature is 873 K in the TGAP system. A detailed description of the TGAP system is provided elsewhere [16]. Two 1000 Torr range capacitance manometers (MKS Instruments) are used to measure the total pressure in the primary side (pP) and the secondary side (sP). Two mass flow controllers (Sierra Instruments) are used to control and monitor the mass flow rates in the primary side (pF) and the secondary side (sF). A 10 cm^3 ion chamber (Tyne Engineering) is used to measure the tritium concentration in the primary side (pIC), and a 1000 cm^3 ion chamber (Tyne Engineering) is used to measure the tritium concentration in the secondary side (sIC). Two additional TCs are located at the ionization chamber locations and are used to measure the gas temperatures in the primary side (pTC) and the secondary side (sTC).

A Tritium Source System (TSS) was constructed to independently control H_2 and T_2 partial pressures in the balance helium (He) gas stream to the primary side. The TSS consists of a pressure vessel (maximum operating pressure at 2.3 × 10^6 Pa) with several mass flow controllers (Sierra Instrument). Prior to mixing, the pressure vessel was baked at 573 K for 24–48 h to remove moisture and outgas residual gas. Pure tritium gas was initially filled into a tritium transfer cylinder (volume 11.35 cm^3) using INL's Tritium Storage and Assay System. The tritium-filled cylinder was transferred to the TSS and the tritium gas was expanded into the TSS volume of 3770.24 cm^3. Maximum allowable inventory in TSS is 25 Ci. A commercially available 1000-ppm H_2-He gas cylinder was used to pressurize the TSS volume to approximately 2.0 × 10^6 Pa. The majority of tritium is converted to HT form in the TSS due to high (1000-ppm) hydrogen concentration. It is important to note that we intended to keep the hydrogen partial pressure always higher than tritium partial pressure to satisfy the $p_{H2} > > p_{HT} > > p_{T2}$ condition and keep majority of tritium in the form of HT. The 1000-ppm H_2-He mixture was also used as a sweep gas in the secondary side to keep the H_2 partial pressure identical in both primary and secondary side. At a total pressure of 100,000 Pa, the 1000-ppm H_2-He mixture provides approximately 100 Pa of H_2 partial pressure.

During a permeation experiment, a 50 standard cubic centimeter per minute (sccm) of H_2-HT-He gas mixture flows from the TSS to the primary side through a molecular sieve trap (pMST) to remove the moisture in the primary gas. Similarly, a 200 sccm of H_2-He sweep gas flows through a molecular sieve trap (sMST) to remove the moisture in the secondary gas. Two moisture sensors (General Electrics) measure the moisture level in the primary (pMOS) and the secondary (sMOS). Tritium containing process gas from the primary and secondary sides is routed to the tritium exhaust processing (TEP) system prior to being exhausted to the stack. The TEP is equipped with 4 catalyst-bubbler sets (3 for secondary, 1 for primary). Each catalyst-bubbler set consists of a catalyst bed (e.g. platinum coated alumina and/or copper oxide) with a resistive heater, and two sets of 30 cm^3 bubblers (e.g. ethylene glycol or water) to convert elemental tritium (HT) to tritiated water (HTO) in a