A Description of the Tritium Facility at the Chalk River Laboratories

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Abstract

AECL's Tritium Facility is located at its Chalk River Laboratories (CRL). The Tritium Facility was originally built to support the tritium technology needs for CANDU reactors and Canadian fusion program. The Tritium Facility commenced its operation in 1979. Since its inception, it has been involved in the development of heavy water detritiation and upgrading processes, development and testing of tritium-breeder materials and design and testing of fusion-fuel cleanup systems for fusion reactor applications, investigation of tritium-materials interactions, tritium storage getters etc. The Tritium Facility also contributed to the design, construction and commissioning activities of the Combined Electrolysis and Catalytic Exchange Upgrading and Detritiation (CECE-UD) Facility at CRL and the Wolsong Tritium Removal Facility (WTRF) in Korea. This paper describes the general set-up of the laboratory, its capabilities and the current tritium-related activities.

1. Introduction

The Tritium Facility is operated by Atomic Energy of Canada Limited (AECL). It is located at the CRL site at Chalk River. The Tritium Facility is a Class A Radioisotope Laboratory and it is listed in the CRL site license as a Class IB Nuclear Facility. It is a unique facility in Canada, and is licensed for handling up to 37 PBq[1] of tritium. The current activities include research and development (R&D) work related to light and heavy water detritiation processes for applications in CANDU and fusion reactors, providing specialized services to CANDU power stations and commercial activities that include supplying tritium, on behalf of Ontario Power Generation Nuclear (OPGN), for use in civilian applications of tritium and tritium gas standards to the industry and R&D laboratories worldwide.

2. Tritium Facility - A Brief Description [1]

The Tritium Facility consists of a set of rooms covering an area of approximately 316 m^2. Various systems for tritium confinement, multiple-barrier containment, atmosphere clean-up and contamination control have been put into place to control the tritium. The "defense-in-depth" principle has been applied to the Tritium Facility and equipment design for tritium handling operations.

The tritium handling equipment is designed to minimize tritium emissions during normal operations and limit transient emissions to the room atmosphere and to the environment. Generally, double-containment-type systems comprising a primary containment system and a secondary enclosure are used. The primary containment is always of a leak-tight design, while the secondary enclosure design depends on the quantity of tritium. The secondary enclosures used in the Tritium Facility

[1] 37 GBq = 1 Ci

include fume hoods, air-purged enclosures and inert-atmosphere glove boxes (IAGBs). Typically, tritium-handling operations involving more than 3.7 TBq in the process are carried out in IAGBs, equipped with recirculating atmosphere purification systems, to minimize tritium releases to the environment [2]. Air-purged enclosures and fume hoods are used for operations involving tritium up to 3.7 TBq. For operations involving tritiated water (HTO[2]) in fume hoods, because of its high radiological hazard, the total tritium inventory is limited to less than 370 GBq. In addition, the specific activity of HTO that can be handled in the Tritium Facility is limited to 37 $TBq.L^{-1}$. High specific activity HTO is handled only using IAGBs or air-purged enclosures.

Since elemental tritium (HT[3]) is radiologically less hazardous than its oxide (HTO) by a factor of about 10 000 tritium is retained in its elemental form as much as practical. HT is typically stored as a metal tritides (titanium or uranium) in specially designed storage beds. Tritium gas standards containing HT in inert gases (<<1 atom% tritium) are stored in compressed-gas cylinders. R&D programs also require handling of tritium in other forms, such as tritiated water or tritiated organics. The inventory of these compounds is generally kept to the minimum required for the purposes of the experimental program, and they are stored and handled in a safe manner.

2.1 Primary Containments

2.1.1 Tritium Gas Handling System (TGHS)

The primary containments used in the TF consist of process vessels, tritium-storage beds, piping, valves and fittings, process monitoring equipment (pressure, temperature, flow, tritium activity etc.). A schematic of the tritium gas handling system (TGHS), used in the TF, which represents a typical primary containment system designed for handling high specific activity HT up to pure tritium is shown in Figure 1. The TGHS is located inside the IAGB which is mounted on top of an air-purged enclosure. A primary function of the TGHS is to remove pure elemental tritium (> 99% purity) from Immobilized Tritium Containers (ITCs[4]) supplied from the Darlington Tritium Removal Facility (DTRF), analyse the tritium gas purity and repackage tritium into sales containers (Amersham tritium containers) for OPGN customers. The other functions include supply of tritium for R&D activities in the Facility and for the preparation of gas standards.

The TGHS is a stainless-steel (Type 304) vacuum manifold with various attached components. Swagelok, Cajon weld fittings (tees and crosses) connect the tubing and allow for a number of valves to be connected to the main line to isolate various sections of the vacuum manifold. These valves are typically stainless-steel, all-welded, bellows-sealed type, purchased with butt-weld ends (Swagelok Nupro Series valves).

[2] HTO is used to represent all three isotopic compositions of tritiated water (HTO, DTO, T_2O).
[3] HT is used to represent all three isotopic compositions of elemental tritium (HT, DT and T_2).
[4] Tritium removed from the heavy water from Ontario Power Generation Nuclear (OPGN) stations at the DTRF is stored in stainless steel tritium storage containers, containing titanium metal absorbent, called Immobilized Tritium Containers or ITCs.

The various components associated with the vacuum manifold are connected via demountable, metal-sealed, high-vacuum fittings (Swagelok Cajon VCR Couplings) and isolated by all-welded, bellows-sealed stainless-steel valves. The use of demountable fittings and valves allows for easy removal of the valves if they should fail. The valves are designed for vacuum and pressure applications with a maximum pressure and temperature rating of 6.9 MPa and 480°C respectively.

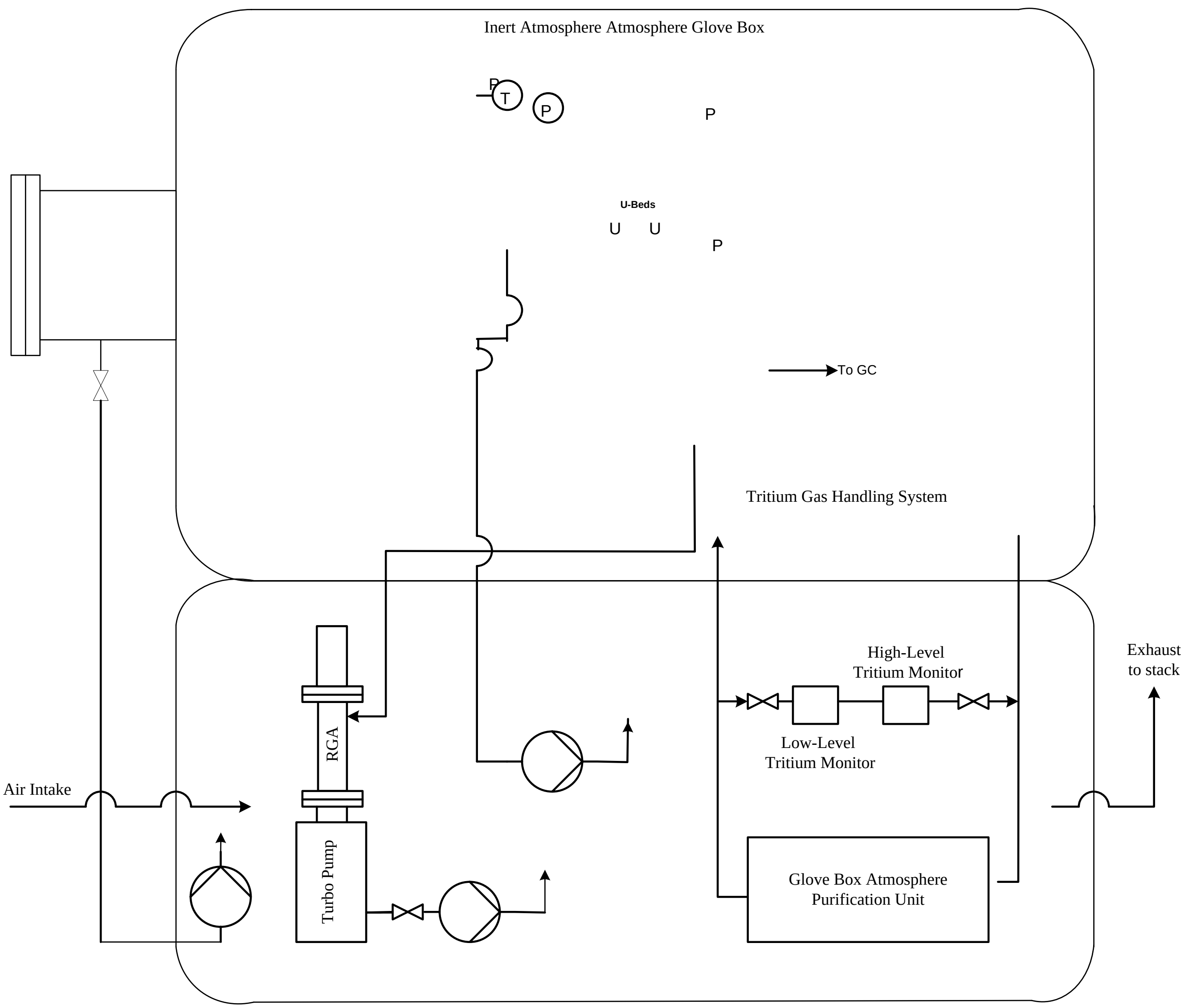

Figure 1 Schematic of the Tritium Gas Handling System and Glove Box

The components attached to the main vacuum manifold include a tritium-compatible, circulating pump (vane pump from Nova Magnetics), all-metal scroll pump (Normetex, 15 $m^3.h^{-1}$) for tritium removal from ITCs, 1-L, 8-L, and 35-L stainless steel expansion vessels, tritium storage beds (titanium and uranium metal getters), and temperature and pressure measuring devices. In addition, a residual gas analyser (RGA), located in the air-purged enclosure, is used for the analysis of tritium in gaseous waste discharges. A Gas Chromatograph, located inside a fume hood, is used for the analysis of purity of tritium gas processed in the TGHS.

2.1.2 Immobilized Tritium Containers (ITCs)

The ITCs are used primarily for shipping tritium from OPGN to CRL. The ITCs are designed as single containment pressure vessels to store up to 18.5 TBq of tritium and to contain tritium decay product, ^{3}He, up to 150 years. The ITCs are made of Type 316L stainless-steel and contain titanium sponge to store tritium as titanium tritide. Two penetrations are welded into the top of the vessel to allow circulation of the gas through the titanium sponge. Porous, 20 μm, sintered, 316 stainless-steel filters are installed in both inlet and outlet tubes to limit tritide particle migration. The inlet and outlet of the bed are fitted with Hoke stainless-steel, bellows-sealed, valves and Cajon VCR fittings. The pressure range for these valves is 1.3×10^{-3} Pa to 13.8 MPa, and the temperature range is from -195°C to 632°C.

The ITCs are originally designed for long-term storage of tritium removed from CANDU reactors. Titanium metal is used as the storage medium because of the extremely low equilibrium partial pressure of tritium over titanium tritide at room temperature [3]. The ITCs are not designed for tritium recovery. Recovery of tritium from ITCs is difficult requiring temperatures in excess of 700°C and long pumping times. High-temperature tritium recovery operations lead to significant contamination of the outside surfaces of the ITCs due to tritium permeation through metal. As a consequence used ITCs are handled as active waste after a single use.

2.1.3 Uranium Storage Vessels

Three types of tritium storage containers based on uranium are used in the Tritium Facility. They are: Tritium-Extraction-Plant (TEP) U-beds, CRL U-beds and Amersham U-beds.

The TEP U-beds were originally designed for use in the CRL Tritium Extraction Plant (TEP). They are used in the Tritium Facility to store tritium removed from ITCs. Two TEP U-beds are used in the TGHS. The TEP U-bed is a single-containment design, fabricated from Type 316 L stainless-steel. The inlet and outlet tubes are fitted with 1 μm sintered stainless-steel filters to limit the escape of uranium fines. The vessels are fitted with Nupro, all-metal, bellows-sealed, valves and Cajon VCR couplings. The TEP U-beds are designed to store up to 8.5 PBq of tritium per storage vessel.

Two CRL U-beds are used for temporary storage of tritiated gas during manipulations in the TGHS, to remove tritium from residual gases in the TGHS before venting to the contaminated stack and for storage of isotopically downgraded tritium resulting from operations in the TGHS. The CRL U-bed is a single-containment design, fabricated from Type 304 stainless-steel. The inlet and outlet tubes are fitted with 1 μm filters to prevent uranium fines from escaping the bed. The vessels are fitted with Nupro, all-metal, bellows-sealed, valves and Cajon VCR couplings. The CRL U beds are designed to hold up to 1 PBq of tritium per storage vessel.

The Amersham U-beds are used for the shipment of tritium to and from CRL to OPGN customers. The Amersham U-beds use a single-containment design. Two types of Amersham U-beds (Mk III and Mk IV) are used. The Mk III and Mk IV are licensed for 2 and 4 PBq of tritium respectively. The design pressure and temperature are 4 MPa and 450°C respectively. The design values are based on a fully loaded bed stored up to 12 months. The design includes a stainless-steel filter to limit loss of uranium fines. The Amersham U-beds are fitted with inlet/outlet Nupro, all-metal, bellows-sealed, valves and Cajon VCR fittings.

Tritium stored in uranium metal can be removed at relatively low temperatures compared to tritium stored in titanium [3]. An equilibrium tritium pressure of 101.3 kPa is reached at a temperature of 432°C [4] for uranium tritide and the equilibrium pressure remains relatively constant over a wide range of tritium-to-uranium atom ratios.

2.2 Secondary Enclosures

The primary containers are enclosed inside secondary enclosures as a second barrier of protection. The type, size, shape, leak rate and the quality of the secondary enclosures generally depend on the total amount and specific activity of tritium in the primary containments and the type of operations performed. Fume hoods, air-purged enclosures and glove boxes and are used in the TF to provide secondary enclosures for tritium handling operations.

2.2.1 Glove Boxes

Two IAGBs are used in the Tritium Facility. One IAGB is used to house the TGHS as shown in Figure 2. The other IAGB is used to house a R&D test facility (See section 3.2.1). The 2.5-m^3 glove box is constructed from stainless steel with polycarbonate windows along the front, top and back. The atmosphere in the box is argon. The glove box atmosphere is continuously monitored for oxygen, tritium and water vapor and purified by recirculation through the glove-box purification system (See Figure 1). The glove box is operated at a slightly positive pressure (250-500 Pa (gauge)) to reduce leakage of air into the enclosure. Pressure control is automatic, using differential pressure switches to control the addition and venting of the argon gas from the box.

Electrical services and gas-transfer lines are brought into the box through penetrations located on the top frame. Instrument cables exit through hermetically sealed feedthroughs located on a panel at the back of the glove box and through feedthroughs installed in the rear window. Readout displays of all instruments are located outside the glove box in instrument racks.

2.2.1.1 **Glove Box Atmosphere Purification System**

There are two gas-purification systems available to the IAGB containing the TGHS. The first is a commercial GPU-20X Inert Gas Purifier from Stainless Steel Equipment Co. The argon atmosphere of the glove box is continuously circulated through the titanium-metal getter in the GPU-20X to remove oxygen, nitrogen, water vapor, hydrogen and tritium from the glove-box atmosphere. The GPU is excellent in maintaining impurities such as oxygen (<1 ppm) and water vapor (<50 ppm) at low levels. The hydrogen and tritium removal efficiency of the GPU is poor and the hydrogen and tritium levels in the IAGB vary with the glove box operations and the performance of the purification system. Consequently, a second purification system (auxiliary cleanup system), based on SAES St707 getter, is used to limit the tritium concentration in the glove box below 3.7 $GBq.m^{-3}$ from both chronic and acute tritium releases. The auxiliary cleanup system is maintained in standby mode with the getter bed at the operating temperature (400°C). The glove-box atmosphere is circulated through the auxiliary cleanup system, automatically, when the tritium concentration in the glove box exceeds 3.7 $GBq.m^{-3}$.

Figure 2 Inert Atmosphere Glove Box

2.2.1.2 **Glove Box Atmosphere Monitoring Equipment**

Oxygen, moisture and tritium in the IAGB are monitored continuously. Continuous monitoring provides timely warning of any leaks in the box structure, malfunctioning of the purification system, or tritium releases from the TGHS or other components into the glove-box atmosphere.

The oxygen and moisture analyzers used have sensors mounted in the glove box. Oxygen analysis is provided by a Teledyne Model 317X Trace Oxygen Analyzer and moisture by a Panametrics Series 4 Hygrometer or equivalent instruments.

One low-level (0-370 $GBq{\cdot}m^{-3}$) and one high-level (0-1 $PBq{\cdot}m^{-3}$), ionization-chamber-based, online, tritium monitors from Tyne Engineering are used to sample the GPU-20X purifier exhaust. These monitors can also be selected to monitor the transfer port during an equipment transfer, if required.

2.2.1.3 **Analytical Equipment**

A RGA (MKS quadrupole mass spectrometer) is used to analyze the impurities and the tritium content in residual gaseous waste in the TGHS, following tritium operations before discharging to the contaminated stack, to ensure that the residual tritium activity in gaseous waste discharges are kept to a minimum. The RGA and the pumping system are located inside the air-purged enclosure below the IAGB (Figure 1).

The hydrogen-isotope Gas Chromatograph (GC) is used to determine the composition (^3He, H_2, HD, HT, D_2, DT, T_2) of the tritium gas removed from storage containers quantitatively. The GC is located in a fume hood near the IAGB. The GC is connected to the TGHS in the IAGB via carrier gas lines (Figure 1). The GC model used is a Carle Series 100 AGC, with a thermistor-type detector. The GC separation column is a packed column of FeOH-coated alumina, cooled to liquid N_2 temperature to achieve separation of all six isotopes of hydrogen and ^3He. The carrier gas used is neon at a flow rate of 15 $mL{\cdot}min^{-1}$. A 3-mm O.D., 1.5-m long GC column is used, resulting in an analysis time of ~30 min for a sample containing all six isotopes.

Two liquid scintillation counters (LSCs) are used in the Tritium Facility (Wallac, Winspectral, 1414 and Perkin Elmer, TriCarb 2810TR). The LSCs are used to measure tritium surface contamination in the Tritium Facility using wipes and for assaying tritium gas standards.

2.3 Tritium-in-Air Monitoring System

Real-time, ionization-chamber-based, fixed, tritium-in-air monitors (TAMs) are used in the Tritium Facility to monitor the tritium concentration in room air and in the ventilation exhausts. Overhoff tritium monitors are used throughout the Facility. These monitors performed reliably over the years. These monitors are equipped with both visual and audible alarms to indicate any tritium release in a timely manner. The response to tritium alarms in the Facility is based on the Derived Air Concentration (DAC) of tritium in air. Although the major radionuclide handled in the Tritium Facility is HT, the DAC value for the Tritium Facility is based on HTO vapor, which is radiologically more toxic. While the DAC value provides an estimate of the committed dose to an exposed worker, an accurate dose to an exposed worker is determined by measuring the tritium content in urine samples (bioassay). For work in the Tritium Facility, the "working" DAC value is established conservatively as 370 $kBq{\cdot}m^{-3}$. The DAC for HTO (DAC_{HTO}) is defined as that concentration of airborne tritium activity that would result in the intake of ALI (Annual Limit on Intake for HTO) by a worker exposed continuously for one year (2 000 working hours) at a breathing rate of 1.2 $m^3.h^{-1}$ [5]. The ALI for HTO (1 GBq) is defined as the derived limit for the amount of tritium taken into the body of an adult worker, based on reference man, by inhalation and/or ingestion with no external exposure that would result in the committed effective dose limit of 20 $mSv.a^{-1}$ [5]. The calculated DAC values for HTO (DAC_{HTO}) also accounts for the exposure due to absorption of HTO through the skin (1/3 of the exposure is due to skin absorption).

2.4 Operating Experience

The Tritium Facility has been operating safely for over 30 years. No major tritium compatibility issues associated with equipment and components used in the Tritium Facility were encountered over the life time of the Facility other than age-related failures.

2.4.1 Tritium Gas Handling System

2.4.1.1 Tritium Storage Beds

Over the years a large number of ITCs have been processed in the Tritium Facility. The ITCs are used only once and the only problem encountered with ITCs was a cracked weld in one of the ITCs that was found during an inspection.

From 1991 to 2011, approximately 650 tritium shipments have been made using an original stock of 19 Mk III and 21 Mk IV Amersham U-beds. Mk IV beds are used more frequently than Mk III beds. At the end of 2011, a total of 18 Mk III and 8 Mk IV beds remained in service. The Amersham U-beds were removed from service due to valve failures. Seat leakage was found to be the dominant valve failure mechanism. Only one Amersham bed was removed from service due to bellows failure.

2.4.1.2 Pumps

The Nova Magnetics vane pump used in the TGHS failed completely after ~9 years in service from 2001 to 2010. The pump is lubricated with ~20 mL of OS-124, polyphenyl ether lubricant. This pump is exposed to pure tritium during its use and pump failure was traced to radiation degradation of the lubricant used in the pump (the lubricant has turned into a gel like consistency). The pump was replaced with an identical spare unit. In order to avoid similar failures, the lubricant needs to be replaced periodically. Replacement of the lubricant in this pump is difficult and requires removal of the pump from the glove box.

All oil lubricated pumps, except for the Nova Magnetic pump, used in the TGHS were replaced recently with dry pumps to avoid the need for oil replacement and handling of tritiated-oil waste. However, there is no dry pump currently available to replace the Nova Magnetic pump.

The previous pump of the same model was removed from service once to replace the transmission, due to a damaged gear. The only other maintenance work involved replacement of carbon brushes in the DC motor due to wear.

2.4.1.3 Valves

Valve failures in the TGHS are rare. The valve in the TGHS, at the attachment point of the Amersham U-beds, is replaced periodically (~every 2-3 years) due to seat failure. This valve is affected by the exposure to high temperatures during conditioning of the Amersham U-beds.

3. Tritium Facility Activities

3.1 Commercial Activities

The main commercial activities carried out in the Tritium Facility include supply of pure tritium (>99%) to the manufacturers of tritium lights, major fusion research facilities and manufacturers of tritium-labelled compounds worldwide under a commercial contract with OPGN and supply of tritium gas standards to CANDU stations and manufacturers of tritium-monitoring equipment for calibration.

3.2 Research and Development Activities

3.2.1 Combined Electrolysis and Catalytic Exchange Process (CECE) for Water Detritiation

AECL has been actively involved in exploring advanced electrolysis technologies for its Combined Electrolysis and Catalytic Exchange (CECE) technology for water detritiation applications [6]. A small-scale, close-cycle, CECE system (mini-CECE) using a proton-exchange-membrane (PEM) type electrolysis cell suitable for service with tritium concentrations up to 37 $TBq.kg^{-1}$ (water) has been designed and built in collaboration with Tyne Engineering. Following extensive testing with non-active heavy water, this system was installed in an inert atmosphere glove box equipped with a dedicated cleanup system. This system is used, as a test facility, for the measurement of membrane performance, over long periods, with tritiated water (~37 $TBq.L^{-1}$). This facility also allows for the monitoring for any effects of membrane degradation products on the performance of exchange and recombiner catalysts in the CECE process. A photograph of the mini CECE system, before installation in the glove box, is shown in Figure 3.

3.2.2 Testing of PEM Cell Membranes for Tritium Compatibility

The objective of this work is to screen both commercially available and in-house developed membrane materials for tritium compatibility for use in PEM cells in the CECE process for water detritiation applications. The tritium compatibility of selected membrane materials is assessed by exposing the membrane materials to high-specific-activity tritiated water (37 $TBq.kg^{-1}$) and by assessing the degradation characteristics as a function of the exposure time. The tritiated water is prepared in-house by oxidizing elemental tritium over a copper-oxide bed. The experimental system to prepare and transfer tritiated water to membrane exposure vessels is housed in an air-purged enclosure.

3.2.3 Tritium Permeation Characteristics of Fusion and GEN-IV Reactor Materials

Tritium permeability data for candidate alloys considered for structural components in high-temperature-gas-cooled reactors (HTGRs [7]) coupled to hydrogen production processes and fusion reactors are not widely available in the literature. The key objective of this work is to experimentally determine the tritium permeability data for candidate alloys and potential permeation barriers to reduce tritium permeation through these materials, under representative operating conditions.

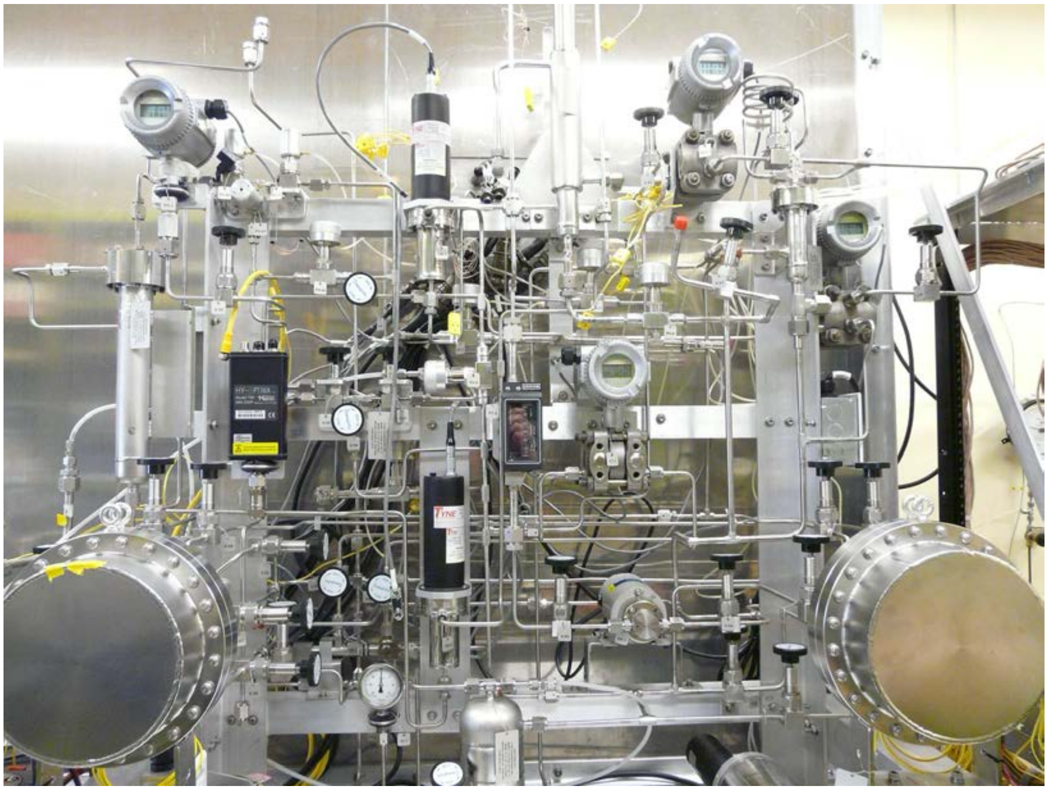

Figure 3 Mini CECE System

In the breeder blanket and the fuel processing systems, in fusion reactors, the structural materials are exposed to non-trivial concentrations of tritium and high temperatures. These conditions promote tritium permeation through structural materials. Some of the key structural materials considered for these applications include modified austenitic stainless steels and reduced activation, ferritic/martensitic (RAFM) steels.

In HTGRs some of the tritium generated in the reactor core will accumulate in the primary He coolant and permeate through the intermediate heat exchangers (IHXs) to the secondary He coolant that carries the heat to the hydrogen production processes [7]. Consequently, tritium generated in the HTGRs can migrate through diffusive/bulk-flow mechanisms and lead to tritium contamination of the hydrogen product, process chemicals, and plant equipment in hydrogen production processes. The measures to reduce or eliminate tritium contamination of the hydrogen production processes include a combination of source-term reduction, tritium removal from the He coolant and use of permeation barriers to reduce tritium permeation through the IHXs. The candidate alloys considered for IHXs are Haynes 230, Inconel 617, and Incoloy 800H

An experimental system was designed and built to measure tritium permeability data, using test samples of candidate alloys, in the temperature region 300 to 950ºC to cover both fusion and HTGR conditions. This system is installed inside an air-purged enclosure in the Tritium Facility.

4. Summary

Brief descriptions of the Tritium Facility located at the CRL site, its tritium-handling capabilities, operating experience and current activities are given. It is a unique facility in Canada, and is licensed for handling up to 37 PBq of tritium. The Tritium Facility has been operating safely with tritium since its inception in 1979. A project is currently underway to build a new Tritium Facility to replace the existing Facility.

5. References

[1] J.M. Miller, W.J. Holtslander, R.E. Johnson, “The Tritium Laboratory”, Fusion Engineering. and Design, 12,349 (1990).

[2] C.L. Gallagher, K.D. McCrimmon, “Operating Experience of the Tritium Laboratory at CRL”, Fusion Science and Technology, 48, 237 (2005).

[3] I. Lewkowicz, in Hydrogen Metal Systems I, Solid State phenomena, F.A. Lewis, A. Aladjem eds. Vols. 49-50, Scitec Publications, 1996.

[4] T. Schober, in Hydrogen Metal Systems I, Solid State phenomena, F.A. Lewis, A. Aladjem eds. Vols. 49-50, Scitec Publications, 1996.

[5] International Atomic Energy Agency (IAEA) Safety Standards Series, Safety Guide, “Assessment of Occupational Exposure due to Intakes of Radionuclides”, No. RS-G-1.2, 1999 and Canadian Nuclear Safety Commission (CNSC) Radiation Safety Data Sheet for tritium, CNSC, 2011.

[6] H. Boniface, S. Suppiah, K. Krishnaswamy, L. Rodrigo, J. Robinson and P. Kwon, “A Small Closed-Cycle Combined Electrolysis and Catalytic Exchange Test System for Water Detritiation”, Fusion Science and Technology, 60, 1347(2011).

[7] R. Elder, R. Allen, “Nuclear Heat for Hydrogen Production: Coupling a Very High/High Temperature Reactor to a Hydrogen Production Plant”, Progress in Nuclear Energy, 51, 500 (2009).