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ATOMIC ENERGY
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L'ÉNERGIE ATOMIQUE
DU CANADA LIMITÉE

EXPERIENCE IN HANDLING CONCENTRATED TRITIUM

Expérience de la manipulation de tritium concentré

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Chalk River, Ontario

December 1985 décembre

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Notes préparées pour une présentation lors du cours d'instruction "Le tritium dans la fusion nucléaire" au Centre de Recherches Nucléaires de Karlsruhe, République Fédérale de l'Allemagne, 1985 Juin 18-21.

Résumé

Ces notes décrivent l'expérience de la manipulation du tritium concentré sous la forme d'hydrogène accumulé au Laboratoire du Tritium des Laboratoires Nucléaires de Chalk River. Les techniques d'opération des boîtes à gants, les systèmes de pompage, les opérations d'hydruration et de déshydruration, et l'analyse du tritium y sont discutés. Des renseignements au sujet de l'Usine d'Extraction du Tritium de Chalk River sont inclus sous forme d'une collection de réimpression des communications présentées à la Rencontre de Dayton sur la Technologie du Tritium, 1985 30 avril-2 mai.

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Notes prepared for presentation at a training course "Tritium In Nuclear Fusion" at Karlsruhe Nuclear Research Centre, Federal Republic of Germany, 1985 June 18-21.

ABSTRACT

The notes describe the experience in handling concentrated tritium in the hydrogen form accumulated in the Chalk River Nuclear Laboratories Tritium Laboratory. The techniques of box operation,pumping systems, hydriding and dehydriding operations, and analysis of tritium are discussed. Information on the Chalk River Tritium Extraction Plant is included as a collection of reprints of papers presented at the Dayton Meeting on Tritium Technology,1985 April 30 - May 2.

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1. CRNL TRITIUM LABORATORY

1.1 Equipment

1.1.1 Inert Atmosphere Glove Box

The major component in the laboratory is the inert atmosphere glove box along with its associated systems (Figure 1). The purpose of the glove box is to provide secondary containment for the tritium containing test apparatus. In the CRNL laboratory tritium is handled in the hydrogen form, and efforts are made to avoid oxidation to the aqueous form. The reason is safety. The aqueous form is about 10^4 times more toxic than tritiated hydrogen (1).

The inert atmosphere glovebox is constructed of 3 mm (1/8") stainless steel sheet (type 304) welded with rounded corners and edges. The windows are made of polycarbonate sheet with the glove ports mounted in the windows. (The location of the glove ports was decided after the equipment has been installed in the box in order to facilitate the maximum access to the apparatus.) The windows are sealed with soft silicone rubber. Window sealing is very important in achieving a low leakage rate for the glove box. The outward leak rate of the CRNL box has been carefully measured using an argon material balance technique to be 0.007% of the box volume per hour with a positive pressure of 125-375 Pa inside the box. The box is operated at a positive pressure to avoid inleakage of oxygen, which could combine with tritium to form tritiated water. Gloves are standard butyl rubber/neoprene multi-polymer gloves to minimize water permeation through the gloves into the box. A commercial stainless steel transfer port is attached to the end of the box to allow movement of equipment in and out of the box without contaminating the box atmosphere with air. The transfer port can be evacuated and purged with argon and the atmosphere monitored for tritium. This ensures tritium is not released to the room when equipment is removed from the box.

The argon gas in the box is continuously purified by circulation through a commercial purifier based on hot titanium metal beds capable of removing of O_2, N_2, H_2O, H_2, CO_x and hydrocarbons. The unit maintains the argon at a few µL/L of O_2 and H_2O. Typical tritium concentrations during operation with concentrated tritium is in the range of 5-20 mCi*/m^3 with excursions to 100 mCi/m^3.

The purity of the argon is monitored by a group of on-line instruments. The oxygen and water are measured by the sensor probes mounted inside the glove box with the signal carried out through hermetically sealed feed-throughs in a panel in the glove box wall. Oxygen is measured with a Teledyne instrument and moisture by a Panametrics hygrometer. Sampling lines to external analysers are also employed for oxygen (Anacon), moisture (Beckman) and tritium (ion chambers). Argon samples can also be taken for external analysis by gas chromatography.

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* 1 Ci = 37 GBq

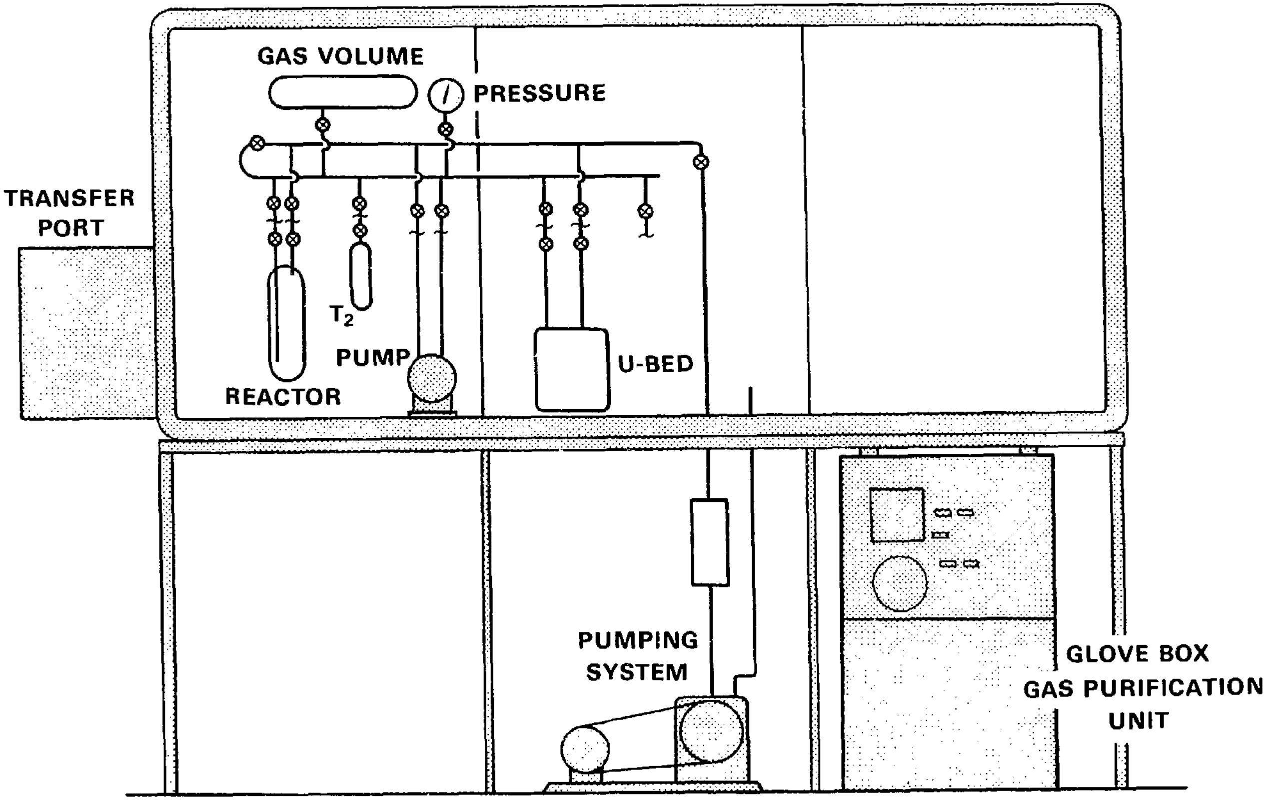

FIGURE 1: CRNL INERT ATMOSPHERE GLOVE BOX

The technique of not placing instruments in the glove box has been followed wherever possible to facilitiate maintenance and to avoid having to move bulky items (which may be tritium contaminated) in and out of the box. To achieve this an instrument sensor feed-through panel, which contains enough feed-throughs for all of the temperature and pressure and other measurements done in the box, has been installed on the back of the enclosure. The integrity of these feed-throughs is very important in maintaining the purity of the argon atmosphere. The ones available from Physicon and Cannon have been found to be satisfactory.

1.1.2 Pumping Systems

The apparatus in the CRNL glove box is a basic vacuum line. It is evacuated using a conventional oil diffusion pump and an oil sealed mechanical vane pump. This pumping system has been found to be quite satisfactory, although it is not generally recommended for tritium service by other people (2). It is economical. In normal operation it is not used to pump tritium; precautions are taken during operation to make sure tritium is removed from the line before it is opened to the pumping system. The criticism of oil-based pumps is that the oil becomes contaminated and handling of this oil is a source of tritium exposure to operating personnel. This has not been the experience in the CRNL Tritium laboratory. Provisions are made on the pumps for removal of the oil by transferring it directly to an evacuated container without contact with the operator. Typical tritium concentrations in the pump oil have been 20-400 mCi/L. The oil used in the diffusion pump is a polyphenyl ether(Monsanto Santovac 5); in the mechanical pump Edwards High Vacuum Oil Grade 16 is used. The mechanical pumps are operated with the minimum amount of oil and oil mist filters plus carbon filters are installed on the discharge from the pumps. Small turbomolecular pumps backed by either a mechanical pump or a cryopump are used for the pumping system on the residual gas analyser in the apparatus.

An oil-free pumping system consisting of an all metal spiral pump (Normetex, France) backed by a metal bellows pump has been recommended by Coffin (3) and installed in a number of tritium applications. Reports on their performance have been good (4). It is important to protect the spiral pump from particulate material with about 5 μm size porous stainless steel filters.

For pumping pure tritium in the system a two stage vane pump, specially designed for tritium service and purchased from Mound Laboratories, is a key component. This is again an oil-sealed pump with a small inventory of 20 mL of the polyphenyl ether lubricant. The advantage of this pump is that it can be used both as a vacuum pump to pump down to approximately 6 Pa or as a compressor to 500 kPa. A reversible motor allows pumping in both directions. The ability to have a significant compression ratio across the pump is a very useful feature in that it allows the transfer of tritium from one vessel to another, which is useful particularly when the amount of tritium is to be measured. Upgraded versions of these two stage vane pumps, with a magnetically coupled drive, are now available commercially from Nova Magnetics Ltd. in Nova Scotia. The magnetic coupling eliminates the need for shaft seal through the pump casing, a potential path for tritium leakage.

A metal bellows or a spiral pump can also be used. The experience in the CRNL laboratory with the metal bellows pump is that pumping stops when the pressure drops below about 300 torr. This is a problem when hydriding tritium in the presence of helium.

1.1.3 Valves and Couplings

All of the valves used in tritium service in the CRNL tritium laboratory are all-metal, welded bellows-sealed valves, primarily Nupro H and B series type valves. Their performance has been very good. Similar type valves manufactured by Hoke have also been used with good success. Valves with soft seats such as copper and organic polymers have been avoided because over tightening can easily damage the seat causing leakage. Organic polymers undergo radiation degradation in the presence of concentrated tritium. In automatically operated valves, soft seats are generally required to obtain an adequate seal. Polyimide seats are reported to be the most stable polymer in tritium service (2). Initially all tritium valves were of the all welded bellows type; however, experience has been such that metal gasketted bellows valves are quite satisfactory. These offer the advantage of ability to replace the stem without having to replace the entire valve.

Couplings have been either butt welded or Cajon VCR. Experience with both have been excellent. The new type of Cajon with a retainer to hold the gasket has been a significant improvement. Without these retainers, it has been important in connections on vertical tubing in the glove box to make sure the female part of the connector was on the lower half of the fitting; otherwise it is difficult to hold all of the components in place and make the connection.

Many valves were purchased as assemblies with couplings on each end. This allows convenient replacement of leaking or failed valves in a system without having to weld. This is particularly convenient for systems contained within glove boxes.

1.1.4 Uranium Beds

Uranium metal is used extensively in many laboratories for short term storage and purification of tritium. There are many designs of uranium beds. When uranium is hydrided and dehydrided it forms a fine metal powder that is pyrophoric in air. The design of the bed must provide for retention of this fine uranium powder, which is a hazard by itself and an even greater hazard if it is in the form of uranium tritide. The retention of the uranium is achieved by the use of porous stainless steel filters (2-5 μm) welded into the inlet and outlet lines of the bed. It is also necessary to provide some means of maintaining the uranium powder dispersed within the bed to avoid a high pressure drop through the bed. This can be accomplished by separating the uranium on porous metal discs within the bed or by dispersal on stainless wire mesh screen. Uranium hydride powder has a lower density than the metal so that it is necessary to provide sufficient void space in the container to accommodate this expansion. High integrity valves such as the bellows-sealed valves should be used on the inlet and outlet lines. In some cases, the uranium bed is designed to have secondary containment, to allow its use outside a glove box. Uranium beds of several designs have been used in the Chalk River Laboratory. They are shown schematically in Figures 2, 3, & 4. Figure 2 illustrates the design of a bed developed from Mound Laboratories in the USA. In this design the uranium is separated with a series of perforated plates. The uranium bed is attached to a flange that provides the top closure for the secondary contain containment vessel. The bed in Figure 3 has been used for the gettering of HT from a helium-3 stream in the NRX reactor. In this bed the uranium is dispersed on a stainless steel knitted mesh. This bed is operated without secondary containment inside a glove box. For transportation between the reactor and the tritium laboratory a secondary sealed container is used. All of these beds have been designed without water cooling. In general, the CRNL Laboratory has avoided the introduction of water lines to the glove box. For the operations carried out, rapid cool down of the bed has not been required.

1.2 Techniques

1.2.1 Hydriding Techniques

Metal hydrides are formed by the direct combination of the metal with hydrogen:

$$M + X/2\ H_2 = MH_X.$$

Hydriding of metals such as uranium, titanium, and zirconium uses similar procedures. In general it is desired to carry out the operation at room temperature to minimize the problem with heat removal from the glove box. There are two main conditions that must be met to achieve reaction at room temperature. The first is to have an active metal surface and the second is to have pure hydrogen or tritium. For titanium and zirconium, it is also necessary to have the metal in a form with high surface area such as sponge or powder. Uranium is too active to start with the metal in a high surface area form and must be initially introduced as a solid bar or similar bulk form. After the initial hydriding which must be done at elevated temperatures (approximately 450°C), the metal is reduced to a fine powder so that subsequent hydridings can be done at room temperature. Titanium and zirconium can be activated by heating in a vacuum, approximately 10-3 Pa, for about one hour at temperatures >400°C. In practice temperatures >500°C are used. The hydrogen must be of high purity. If these two conditions are met then the hydriding will occur quickly at room temperature. This is shown in Figures 5 and 6 taken from data obtained at Chalk River (5).

There is some tolerance of impurities in hydrogen. The effect of helium and oxygen content on the rate of hydriding has been studied in some detail at Chalk River (5,6). It is found that up to 6000 µL/L of oxygen has a minimal effect on the rate of hydriding of titanium sponge at room temperature. Beyond that concentration little reaction with the hydrogen is observed. With helium the commonly observed blanketing effect occurs in a static

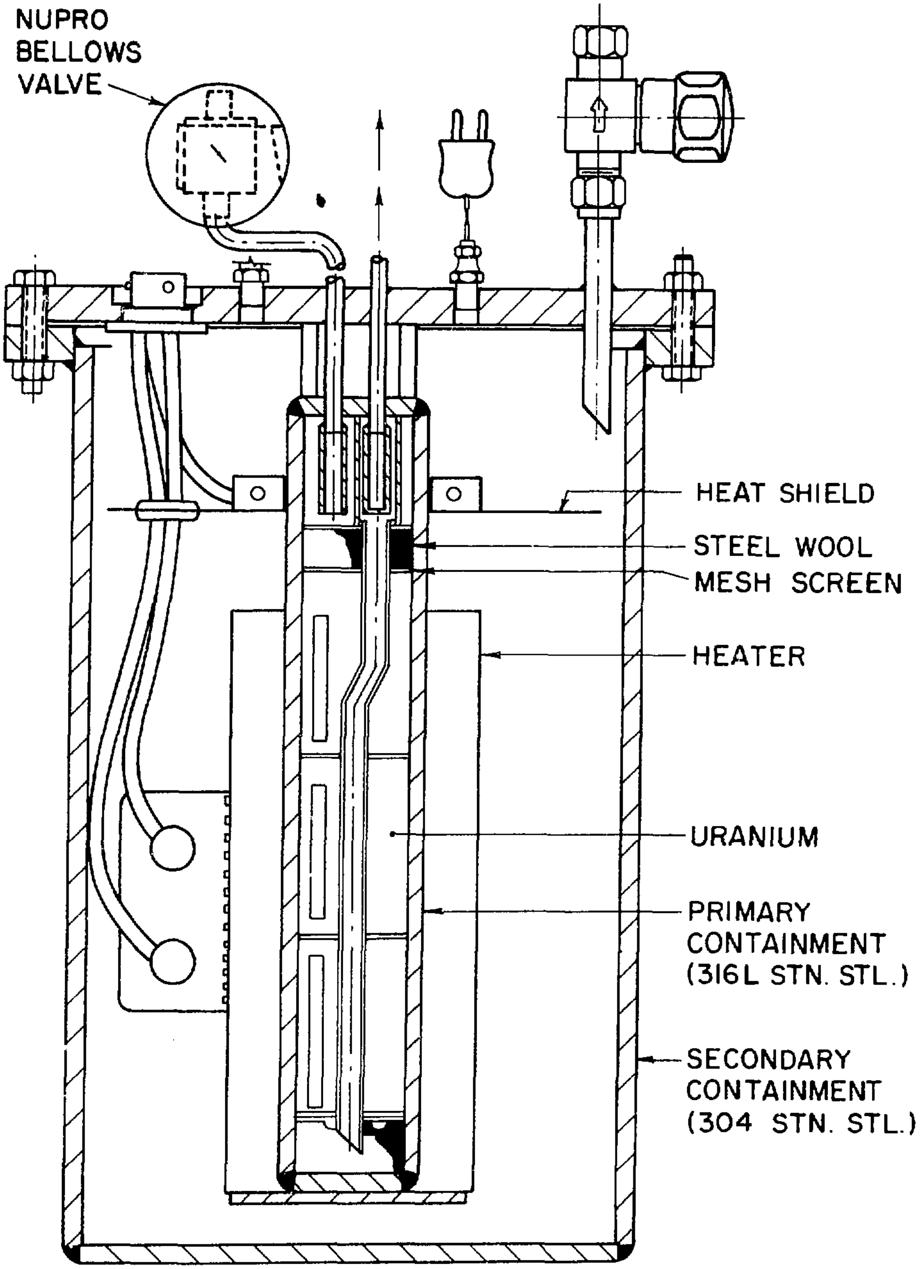

FIGURE 2: MOUND LABORATORIES DESIGNED URANIUM BED

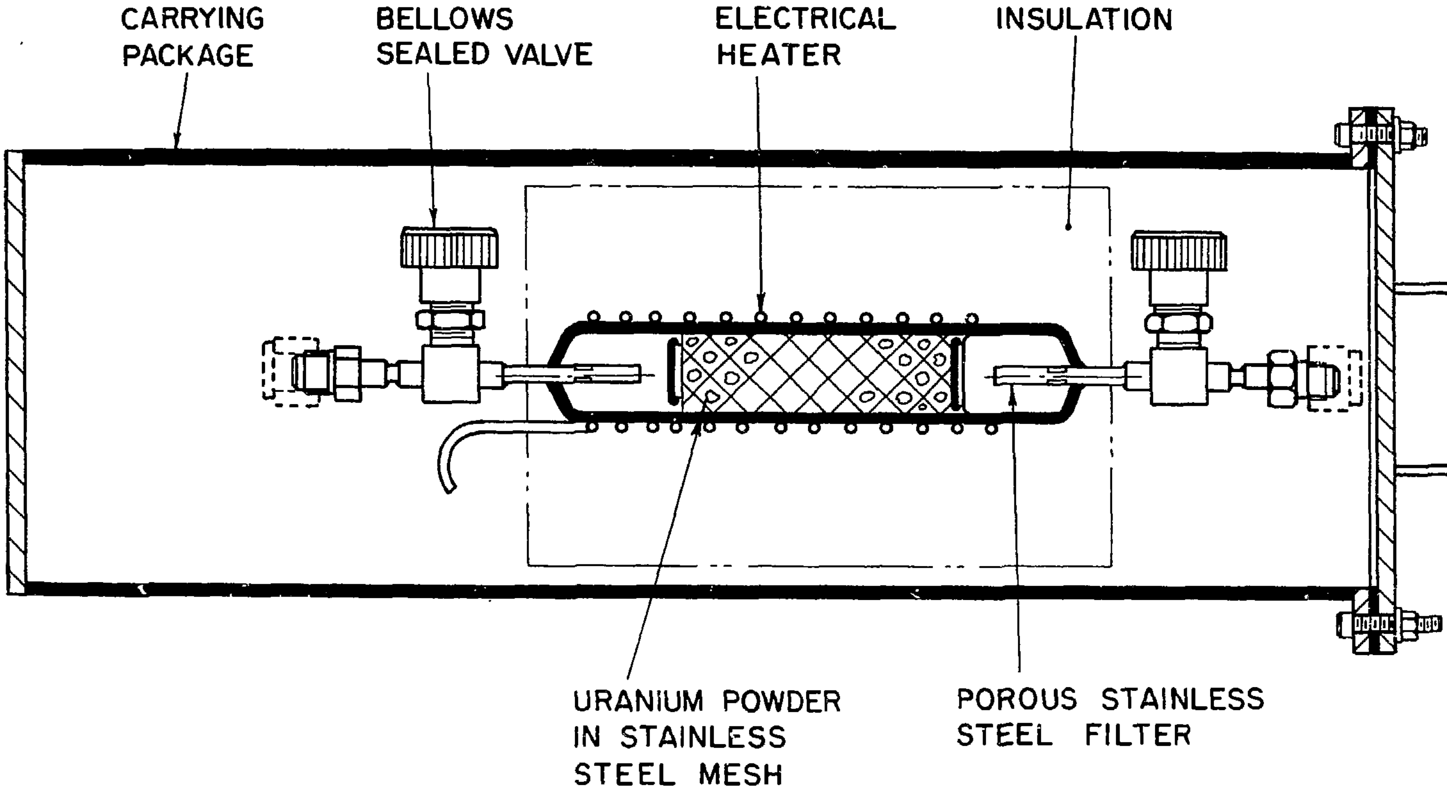

FIGURE 3: CRNL URANIUM TRAP FOR GETTERING HT FROM HELIUM

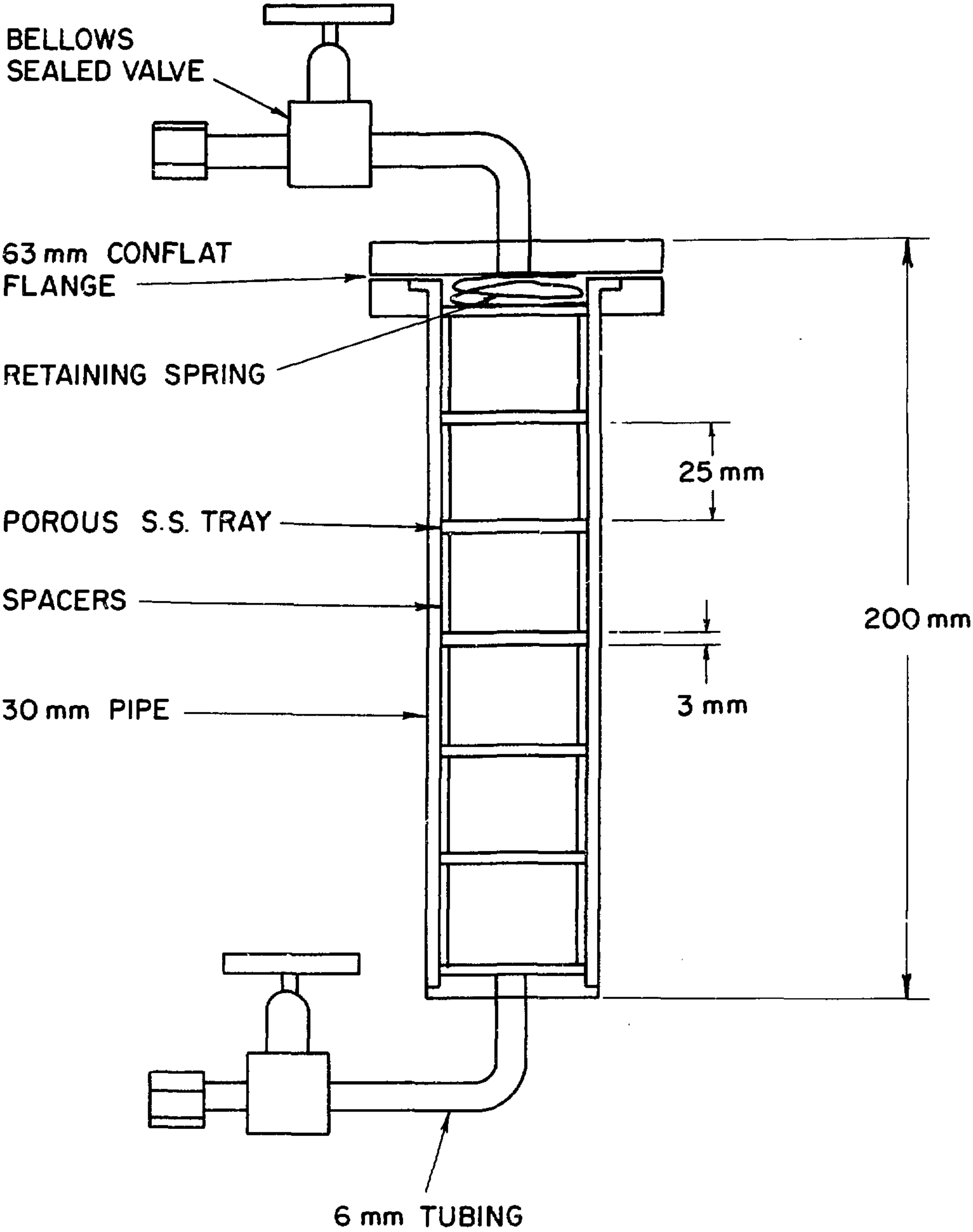

FIGURE 4: CRNL LABORATORY URANIUM BED

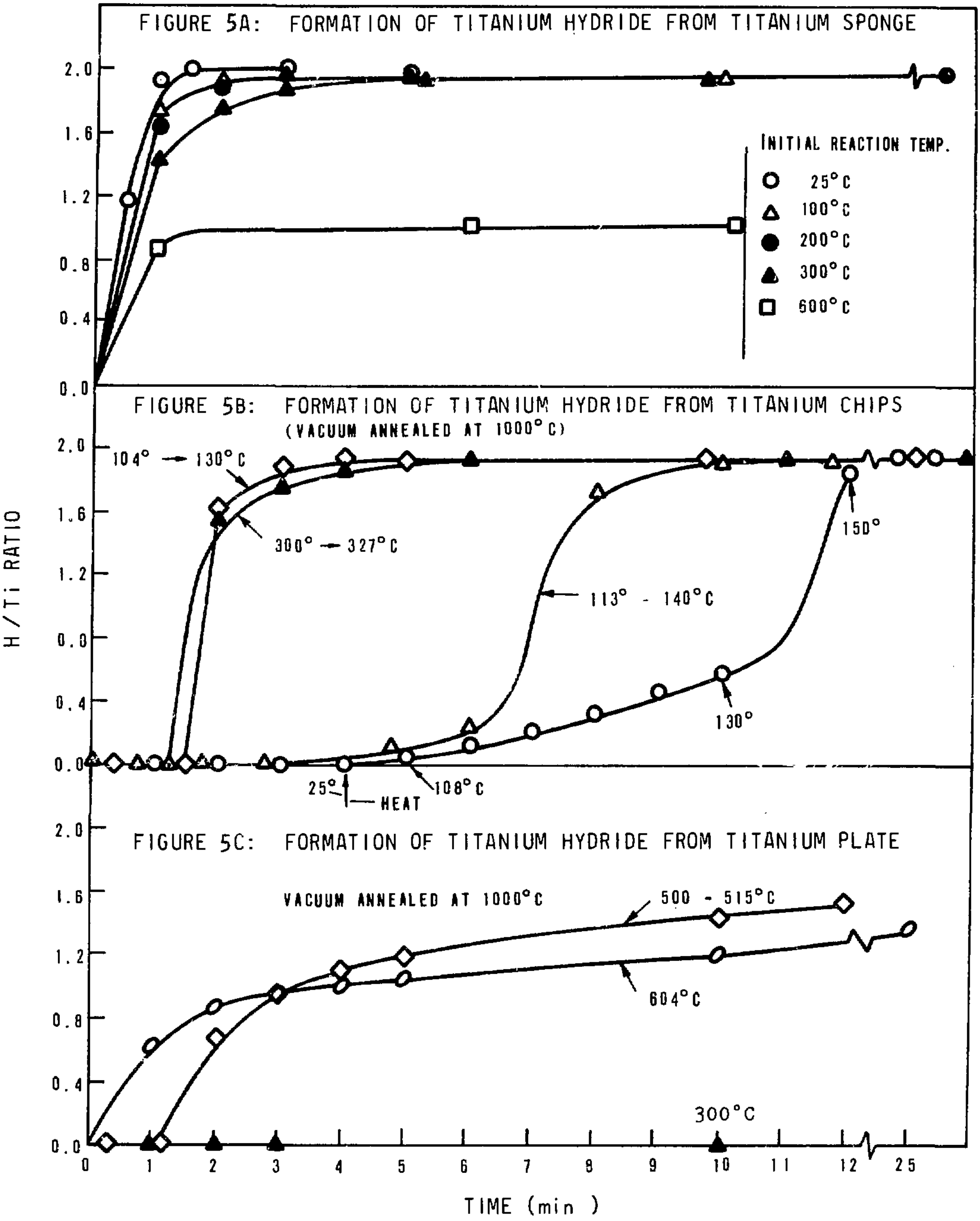

FIGURE 5A: FORMATION OF TITANIUM HYDRIDE FROM TITANIUM SPONGE

FIGURE 5B: FORMATION OF TITANIUM HYDRIDE FROM TITANIUM CHIPS (VACUUM ANNEALED AT 1000° C)

FIGURE 5C: FORMATION OF TITANIUM HYDRIDE FROM TITANIUM PLATE

FIGURE 5: HYDRIDING OF TITANIUM

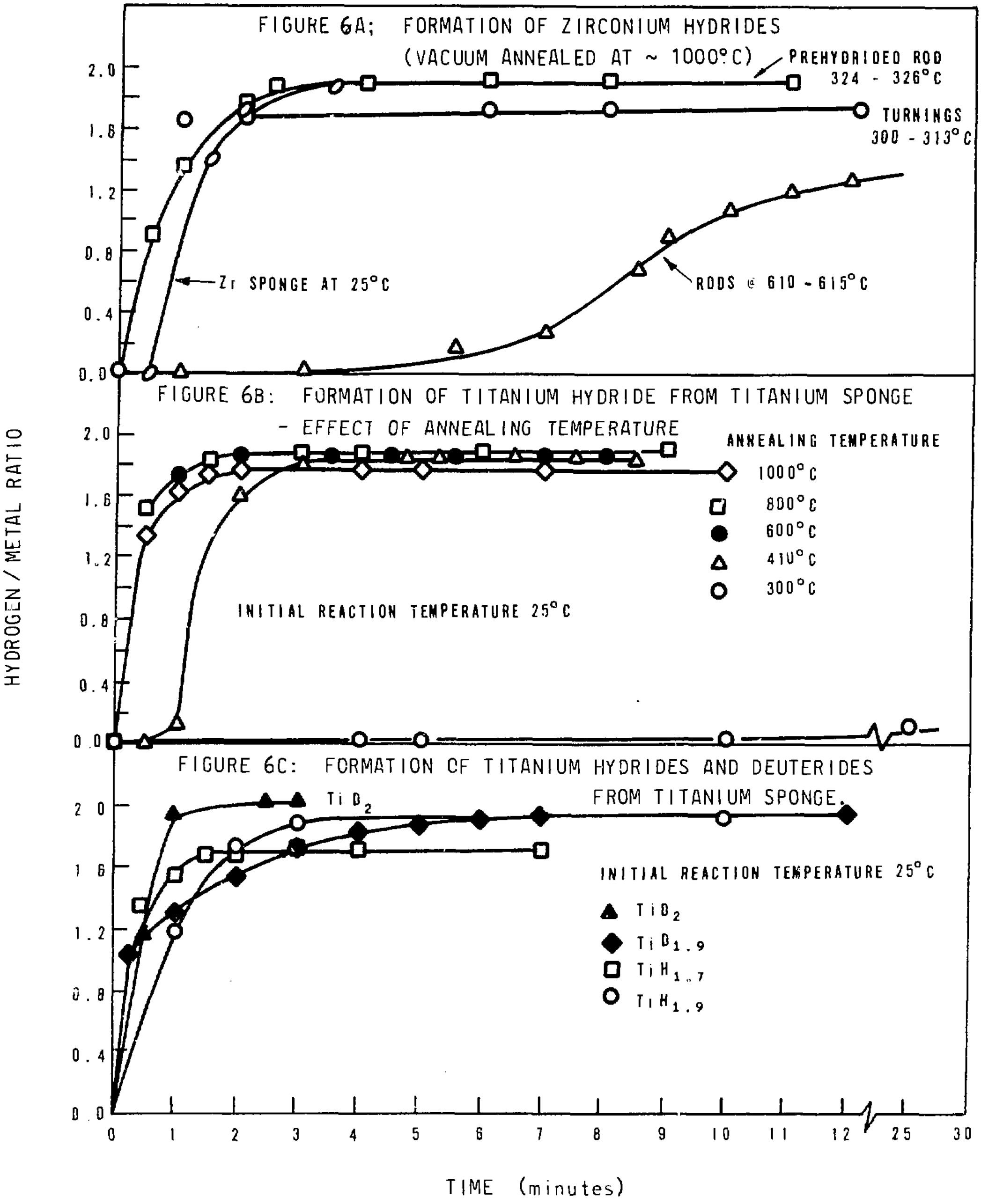

FIGURE 6: HYDRIDING OF TITANIUM AND ZIRCINIUM

system. With concentrations of less than 1% He in the hydrogen in a system where there is no movement of the gas past the metal the reaction starts off normally, but the rate falls off quickly. The hydrogen near the surface reacts leaving an enriched layer of helium-rich gas which provides a barrier to the diffusion of hydrogen from the bulk to the surface of the metal. When the gas is circulated then this stagnent layer of gas is disturbed and the reaction proceeds at the normal rate. These data are shown in Figures 7-12 taken from the work of Miller and Cantlon (6) at CRNL in 1982.

In all of these measurements, the initial temperature of the bed is normally 25°C; however, the reaction is exothermic so that as soon as the reaction with hydrogen begins the temperature in the metal bed rises. The temperature rise depends on the design of the bed, which affects the rate at which heat is conducted away. Some measurements have indicated temperature rises in excess of 100°C in the metal bed. This temperature rise can explain the tolerance to small amounts of impurities such as the observation with oxygen. The higher bed temperature may allow a certain amount of oxide to diffuse into the bulk of the metal and so maintain the surface active.

There are occasions when a normally active metal hydride bed becomes inactive or reacts very slowly with hydrogen. When these occur the standard procedure is to heat the bed to a temperature up to approximately 100°C or until the reaction starts to occur. Once it starts, then it usually quickly achieves a normal reaction rate because of the exothermic reaction. If gentle warming does not start the reaction, the normal practice at Chalk River is to remove the gas and replace it with pure hydrogen and then attempt the hydriding again. This is normally successful. If the gas that would not react is tritium, it is essential to have some place to store it temporarily while the bed is being reactivated. This is why it is important to have at least one spare active uranium bed in the system for temporary storage.

1.2.2 Recovery of Hydrogen (Tritium) from Metal Hydrides

This is a straightforward reversal of the hydriding reaction, and consists of simply heating the metal hydride to generate the desired pressure of hydrogen. The dissociation pressure-temperature characteristics of the uranium-hydrogen system is particularly favourable for the recovery of hydrogen because a pressure of one atmosphere is obtained at approximately 400°C as shown in Figure 13. Even though the permeation of tritium through the stainless steel walls of the metal hydride bed is very small at this temperature, it is normal practice to maintain the pressure in the bed as low as practicable. With the hydrides of titanium and zirconium the corresponding temperatures to obtain a dissociation pressure of one atmosphere are approximately 650 and 900°C, respectively. At these temperatures, permeation through the vessel walls is a problem (7). To minimize permeation during the recovery of tritium from titanium or zirconium tritides it is essential to pump on the high temperature source bed to maintain low pressure. This technique has been demonstrated by Shmayda, Kherani,and Meyer at Ontario Hydro (8). Other approaches have been to allow the tritium to permeate through the first wall and collect it in the secondary containment. A metal getter trap is sometimes used to collect the permeated tritium (2).

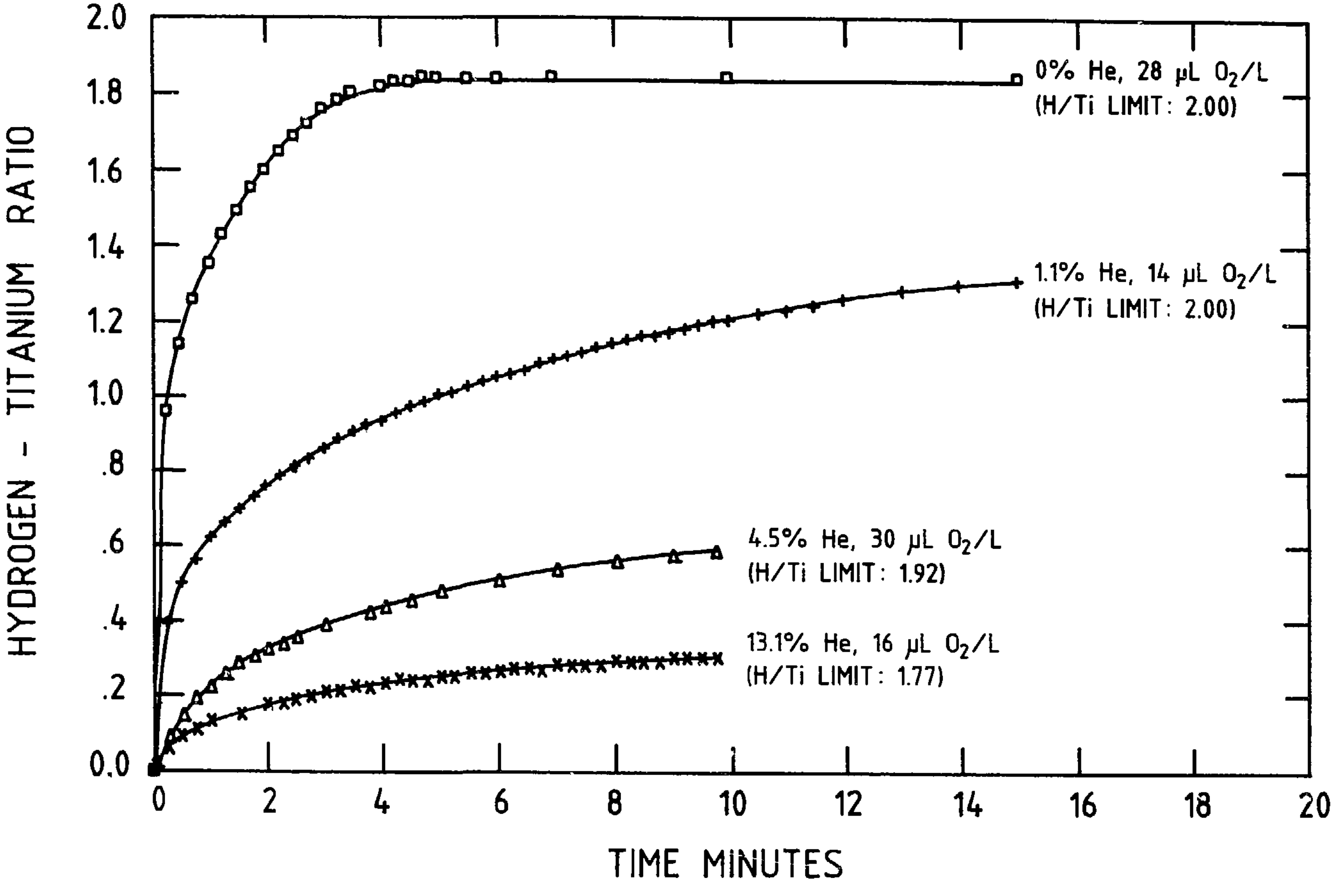

FIGURE 7 STATIC REACTIONS OF H_2 - He WITH TITANIUM SPONGE

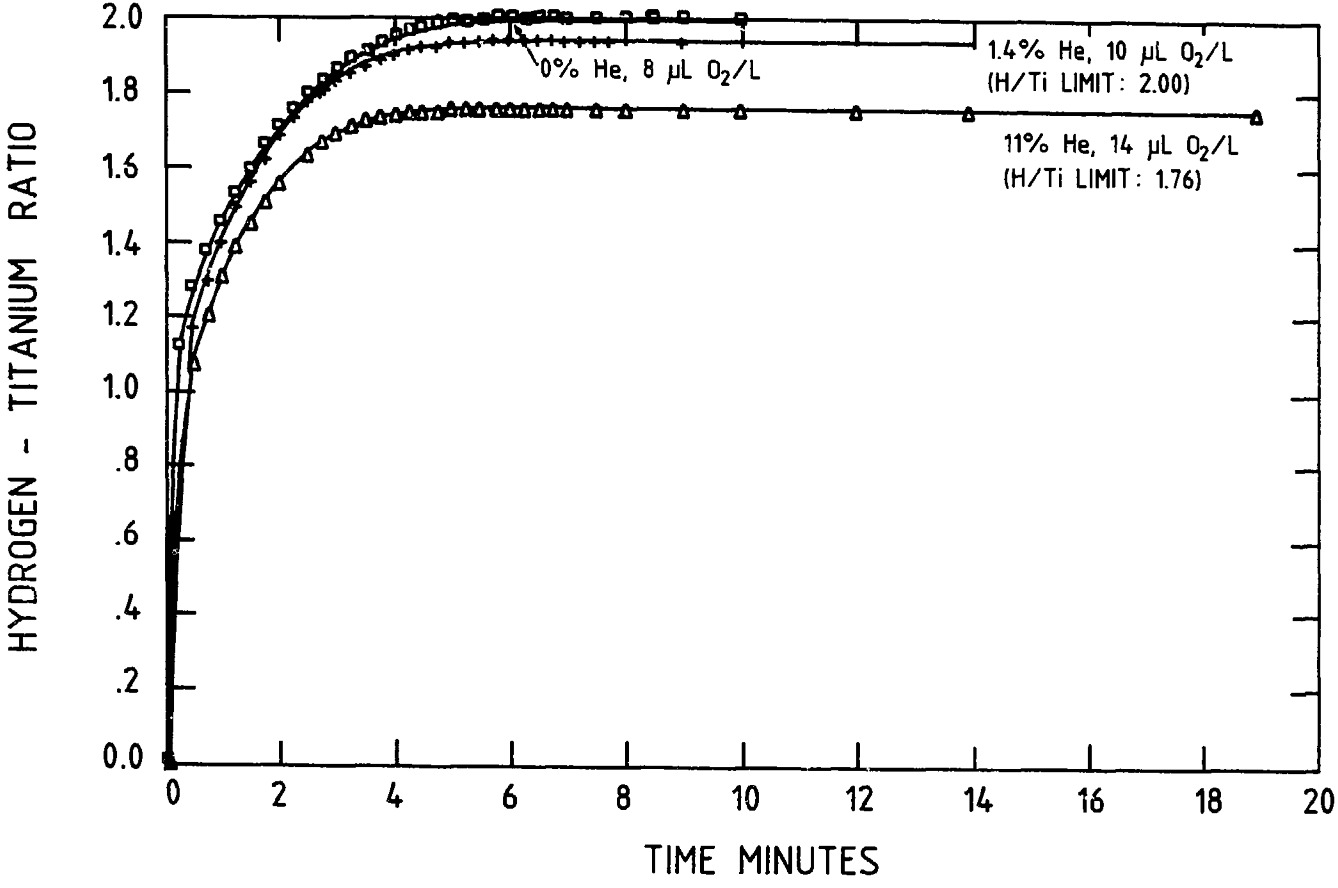

FIGURE 8 DYNAMIC REACTIONS OF H_2 - He WITH TITANIUM SPONGE

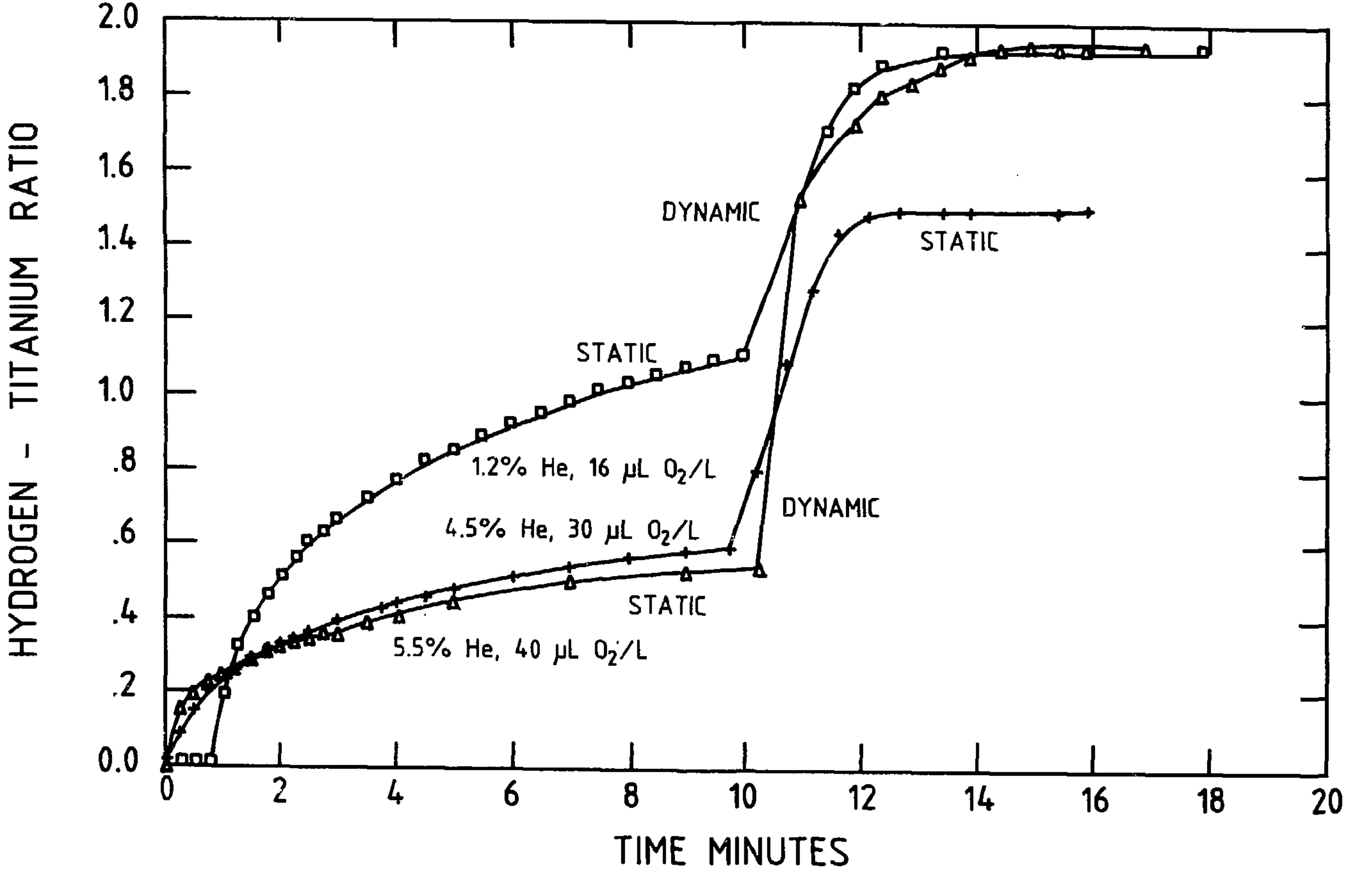

FIGURE 9 EFFECT OF GAS CIRCULATION ON HYDRIDING IN THE PRESENCE OF He

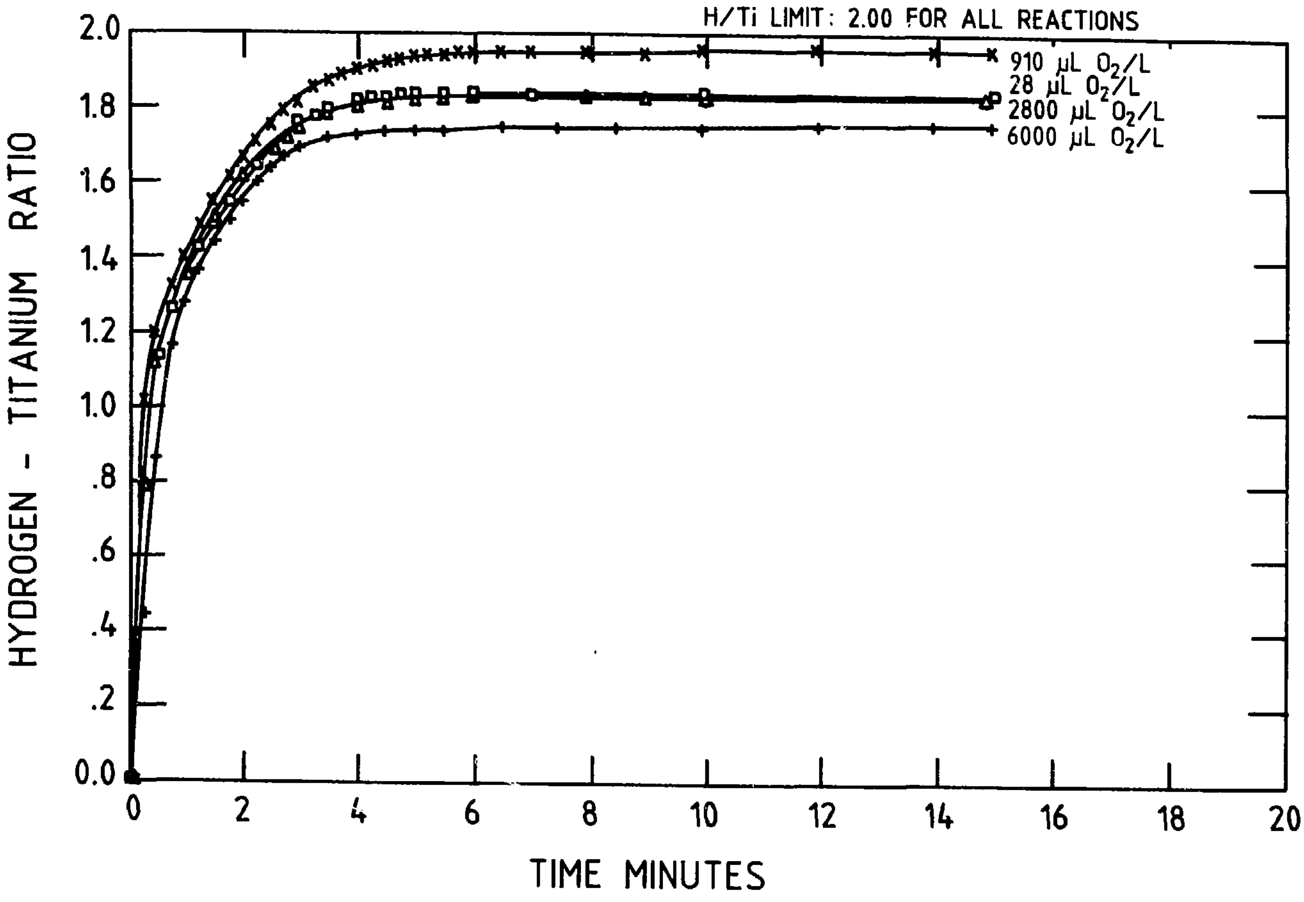

FIGURE 10 EFFECT OF O_2 ON STATIC HYDRIDING OF TITANIUM SPONGE

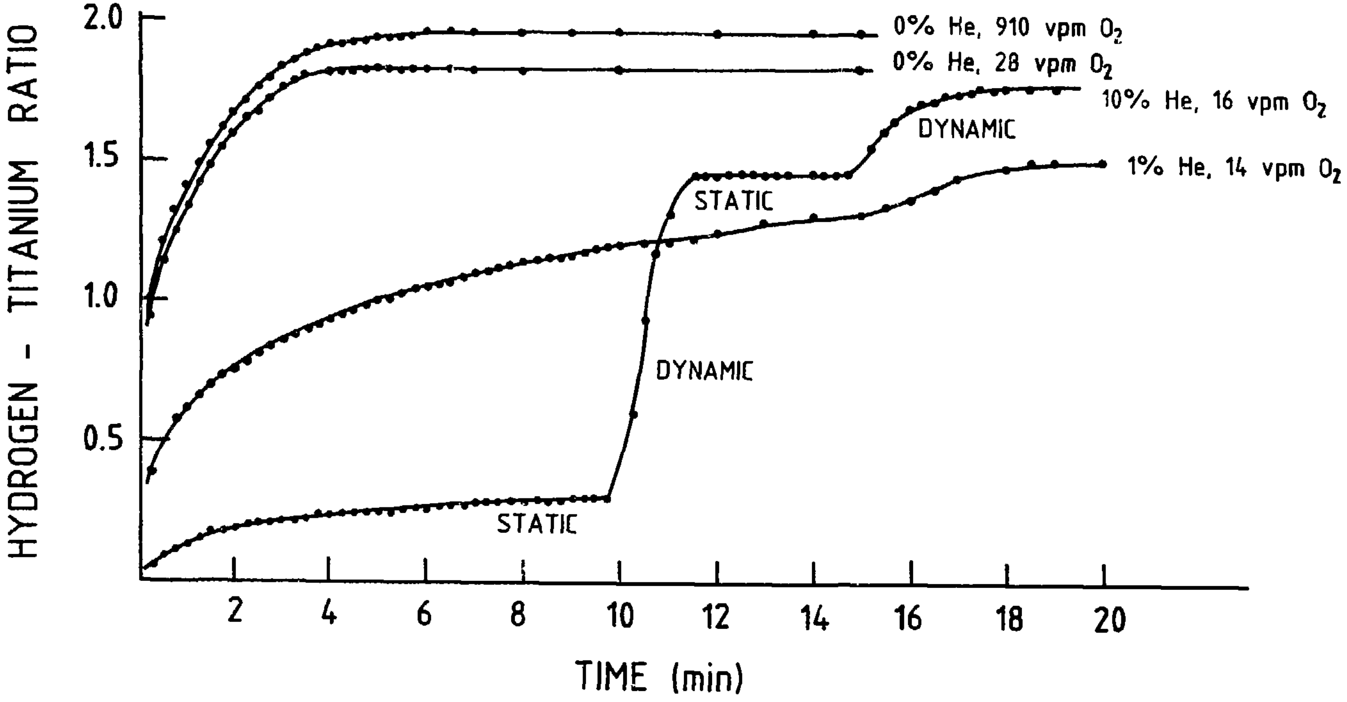

FIGURE 11 EFFECT OF He AND O_2 ON STATIC HYDRIDING OF TITANIUM SPONGE

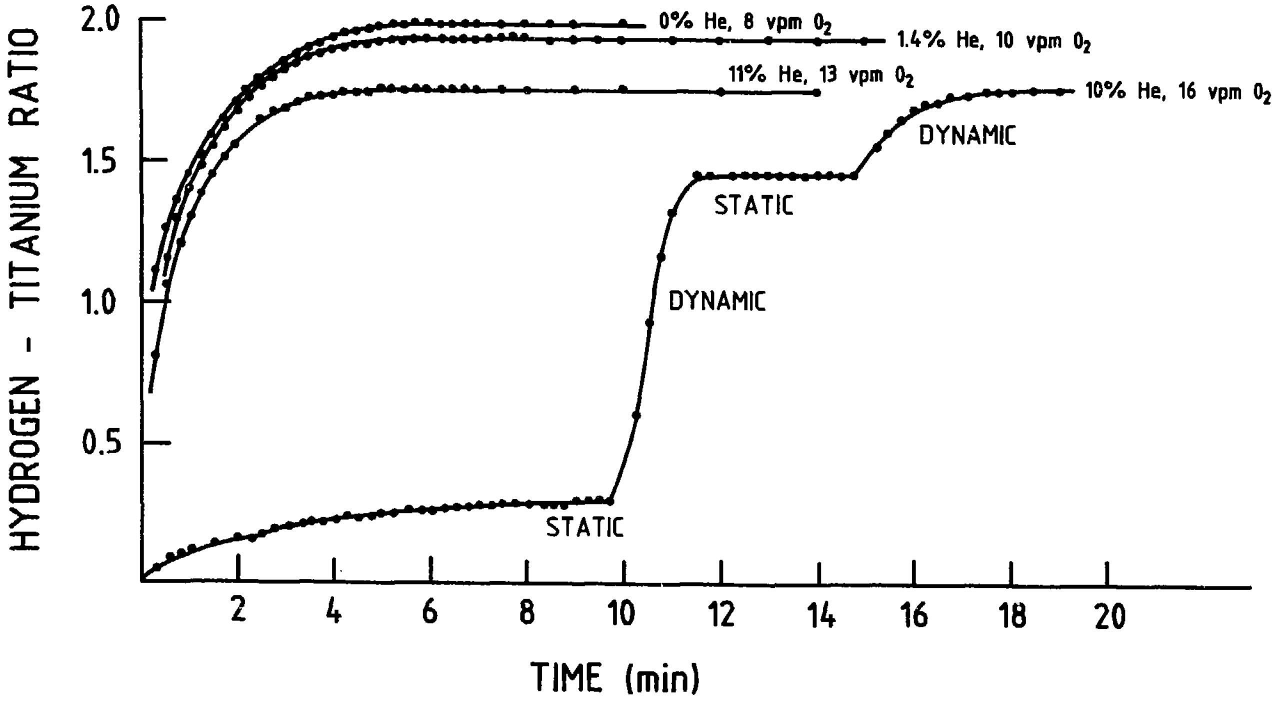

FIGURE 12 EFFECT OF He AND O_2 ON DYNAMIC HYDRIDING OF TITANIUM SPONGE

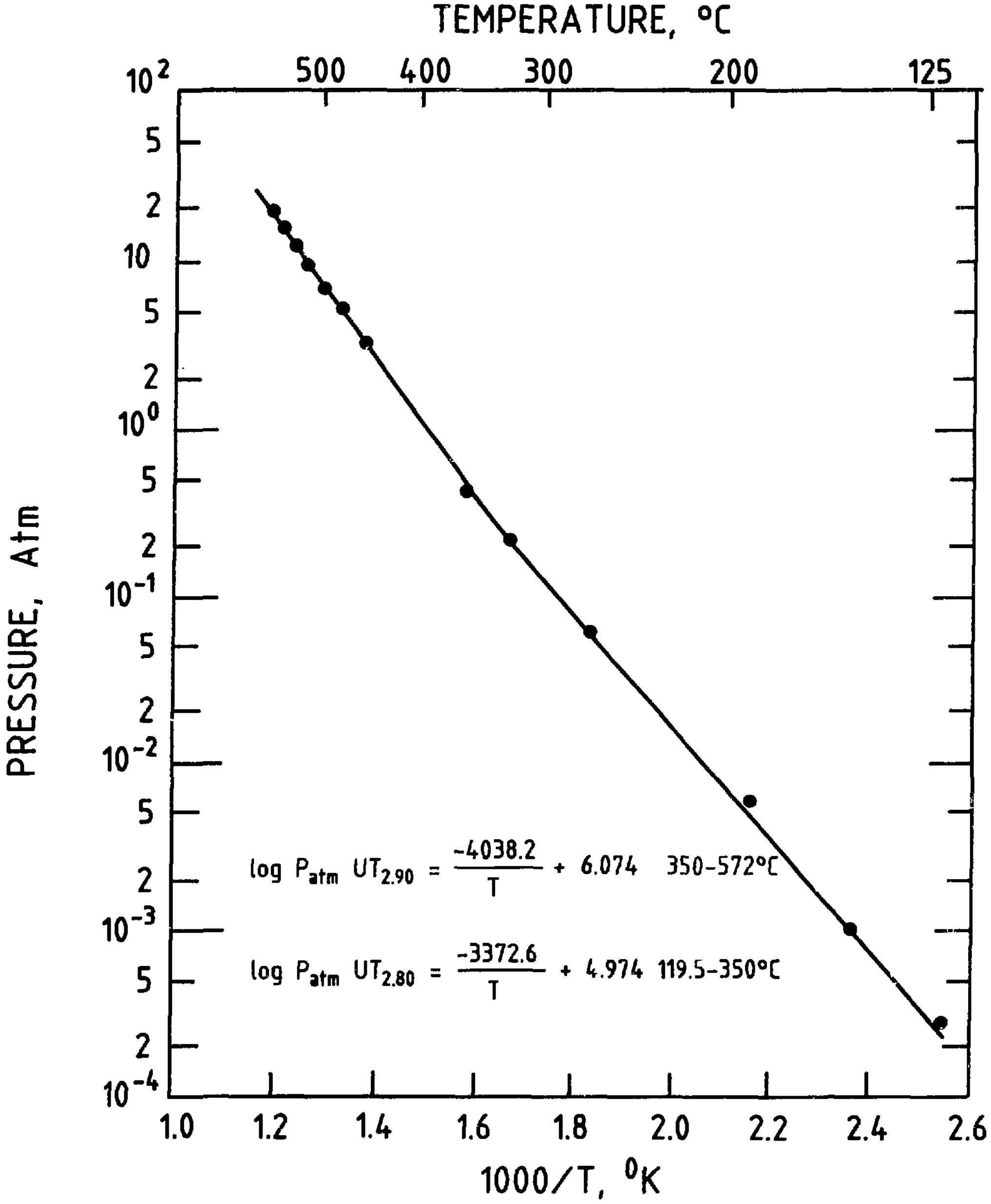

FIGURE 13 THERMODYNAMIC PROPERTIES OF THE URANIUM-TRITIUM SYSTEM (taken from Carlson,B.21)

1.2.3 Isotopic Swamping

The operating philosophy in the CRNL Tritium Laboratory is to avoid releases of tritium whenever possible. After concentrated tritium has been in a vacuum line, residual gas has been removed by storing it either on a uranium bed or elsewhere, there is always a small residual amount of tritium adsorbed on the walls of the equipment. To remove this tritium from the lines the gas line is filled to a few hundred torr with H_2 and the gas circulated through the system; it is then transferred to a uranium bed attached to the line to collect waste which is then tritium much diluted with hydrogen. The procedure is repeated until the inline tritium monitor indicates an acceptably low tritium concentration. This has proved to be an effective technique for minimizing the release of tritium from the laboratory. The procedure generates a certain amount of tritium containing waste. This waste can be either stored or the tritium from it recovered in an isotope separation system such as the CRNL Tritium Extraction Plant.

1.3 Monitoring and Analysis of Tritium

1.3.1 Tritium Monitoring

Tritium monitoring is done exclusively with ion chamber based instrumentation. Two monitors sample the laboratory atmosphere. One is a conventional instrument which measures total tritium and has a remote readout and alarm at the entrance of the laboratory to warn of high tritium levels in the room. The second is a monitor that can discriminate between the aqueous and hydrogen forms of tritium. This is a prototype monitor developed by McElroy and co-workers at CRNL (9) based on a Nafion separator for separating the tritiated water from the air stream and measuring it separately. A commercial version of this instrument is being developed. The effluent from the room and the glove boxes are monitored by two additional instruments.

The glove box argon is monitored by sampling lines from both the inlet and outlet of the argon purifier. The transfer port atmosphere is also sampled and monitored to ensure tritium levels are low enough to open the port to the room. In the event of failure of the primary containment system the tritium concentration would exceed the range of the normal monitors; for this situation an additional ion chamber with an intermediate range is installed in the glove box to monitor the cleanup by the purification system.

Several portable monitors (10) (Model 209, manufactured by Scintrex Ltd. Toronto) are available as required in the laboratory and are used extensively with good results.

Surface contamination of bench and glove box surfaces are monitored by taking swipes of 100 cm^2 areas with filter paper and immersing the swipes in distilled water followed by scintillation counting to detect tritium.

1.3.2 Analysis of Concentrated Tritium

1.3.2.1 Ion Chambers

An ion chamber is a very simple, inexpensive instrument which collects the ions produced by radioactive decay on a central electrode and measures the current produced. The magnitude of the current is dependent on the amount of tritium in the collecting volume. These instruments are used over a wide range of tritium concentrations. For analysis of concentrated tritium, an ion chamber design based on the design published by Carstens and David (12) is used. The chamber is fabricated from a drilled brass rod and has a volume of about 20 mL. The potential to the central electrode is supplied by a 45 volt battery and the current measured by a nA - mA range (Keithley model 160B) digital multimeter. The schematic and the calibration for the chamber taken from reference 12 is shown in Figure 14. When using this technique it is necessary to do the measurement at the same pressure as the calibration or else have a calibration data as a function of pressure. It is the practice at CRNL to either do all of the measurements at 600 torr, and to use the published calibration or operate in the linear region and assume direct proportionality with pressure.

The ion chamber technique is particularly convenient as an on-line device for qualitative analysis of the tritium content of the gas during an operation. It also serves as a qualitative pressure gauge. The disadvantage is that it does not differentiate between the different molecular tritium containing species. It is considered an essential piece of equipment in the Tritium Laboratory.

1.3.2.2 Gas Chromatography

Gas chromatography is the primary technique used at CRNL to determine the isotopic composition of the hydrogen, deuterium, tritium mixtures. The method used is based on the one published by Genty and Schott (12). The column is 1.5 m long packed with ferric hydroxide treated alumina and operated at liquid nitrogen temperature with a neon carrier. The detector is a thermal conductivity type. The gas chromatograph is located outside the glove box, in a fume hood. The gas sampling valve is located in the glove box and the sample is transported to the column by the carrier gas line which has secondary containment between the exit of the glove box and the column in the fume hood. Good separation of all six hydrogen molecular forms and helium is possible. The separation is somewhat variable and changes with the degree of deactivation of the column. When the separation deteriorates, it can be recovered by heating the column for a period of time until the desired separation is achieved. This is somewhat of a trial and error procedure. Good separation stability is observed for periods of the order of a year. The separation is shown in Figure 15. The relative sensitivity factors have been determined by a variety of means. The values obtained are listed in the following table.

Relative Sensitivity Factors for the Hydrogens and the Heliums
on the CRNL G.C.

Species	Relative Response Factor
H_2	1.00
^3He	0.69
HD	0.75
D_2	0.58
HT	0.62
DT	0.41
T_2	0.39

1.3.2.3 Mass Spectrometry

A small residual gas analyser (RGA) is mounted in the glove box and used for isotopic analysis as well as for other impuritiy determination of gas used in the system. It is also used for leak detection. There are two interchangeable heads with different mass ranges of 1-15 amu and 2-100 amu. It has been particularly useful for the analysis of tritiated methanes, which are formed in stainless steel systems exposed to tritium.

The resolution is not sufficient to separate close masses such as HD and ^3He, but it is adequate for most purposes. Used in combination with a uranium bed the relative amounts of HD and ^3He can be determined by adsorbing the HD on the uranium bed and measuring the remaining ^3He on the RGA. The head and the variable leak valve are located in the glove box with the electrical signal taken out through a feed-through to the electronics mounted in an instrument rack located close by. The pumping system consisting of a turbomolecular pump backed by either a cryopump or a conventional mechanical pump is located in a lower air atmosphere glove box under the main inert atmosphere enclosure.

1.3.3 Scintillation Counting

Scintillation counting is used for the determination of tritium concentration in liquids; in the CRNL Laboratory this is primarily in oil from pumps and in water. Water samples arise from the intentional oxidation of a hydrogen stream containing small amounts of tritium as a technique for determining tritium concentration in the gas. The ability of scintillation counting to detect extremely small amounts of tritium makes it particularly suitable for analyzing samples to obtain health physics type data.

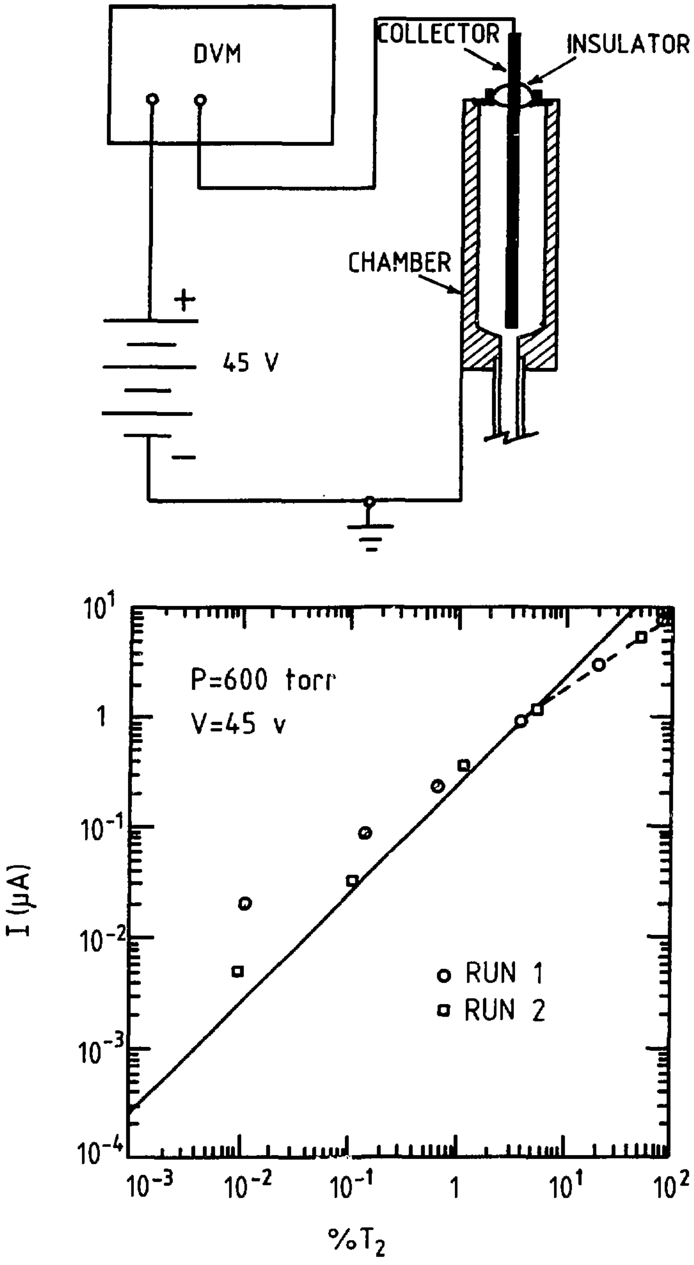

FIGURE 14 LOS ALAMOS HIGH RANGE ION CHAMBER (taken from Carstens and David (11))

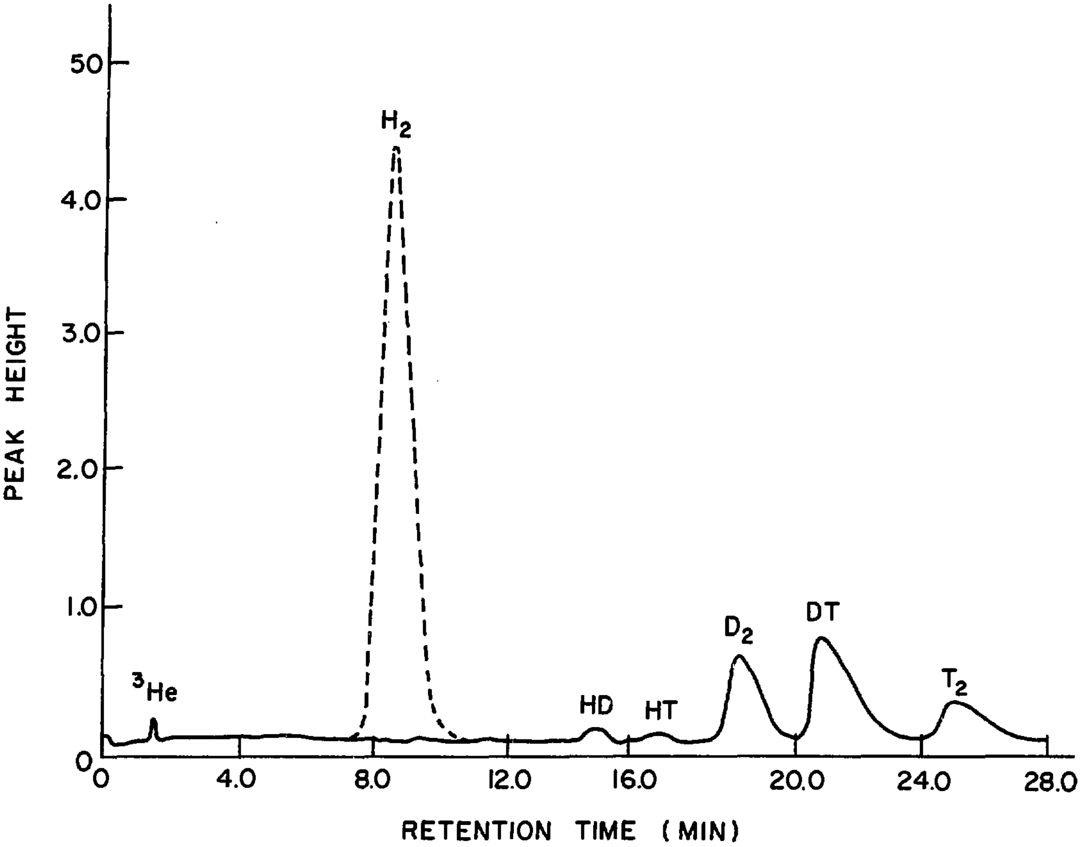

FIG. 15 GAS CHROMATOGRAPHIC SEPARATION OF HYDROGEN ISOTOPES

1.4 Release of Tritium

Tritium can be released either by normal ventilation from the glove box, or by failure of the glove box which provides secondary containment. This tritium is either released directly to the ventilation duct in the case of normal venting or to the laboratory atmosphere in the event of a failure of both primary and secondary containment. Tritium released to the laboratory atmosphere is carried to the room ventilation system via the fume hood and exhaust ducts. The laboratory is not equipped with an air detritiation system. The safety analysis considered the effect of the total release of the entire inventory on a person breathing air at the outlet of the ventilation stack. This was found to give a predicted dose of 9 mrem*.

The release of tritium to the ventilation stack is measured with ion chambers and water bubblers sampling the total stack flow and a separate ion chamber sampling the ventilation flow from the glove boxes. The weekly release limit for the building in which the laboratory is located is 10 Ci.

1.5 Accounting of Tritium Inventory

A computer based system (13) has been developed to record where the tritium in the laboratory is located at any time. The possible locations are a number of metal hydride beds used for experimental or storage purposes. Whenever a transfer between beds is made the amount transferred and its composition is recorded in the computer system. At any time the computer can be instructed to provide a summary of the amount and composition of tritium contained in each bed. The program also calculates the decay of tritium and adjusts the amounts automatically. A copy of the output is shown in Figure 16.

Periodic physical inventories are done where the entire amount of gas on a bed is desorbed and its composition measured. These data are fed into the program to ensure any discrepancies in the quantities and the locations can be identified and examined.

2. CHALK RIVER TRITIUM EXTRACTION PLANT

The features and technology of this plant which is currently under construction at CRNL has been recently described in three papers presented to the American Nuclear Society-Second National Topical Meeting on Tritium Technology in Fission,Fusion and Isotopic Applications, held in Dayton Ohio, April 30-May 2, 1985. Copies of these papers are included as appendices.

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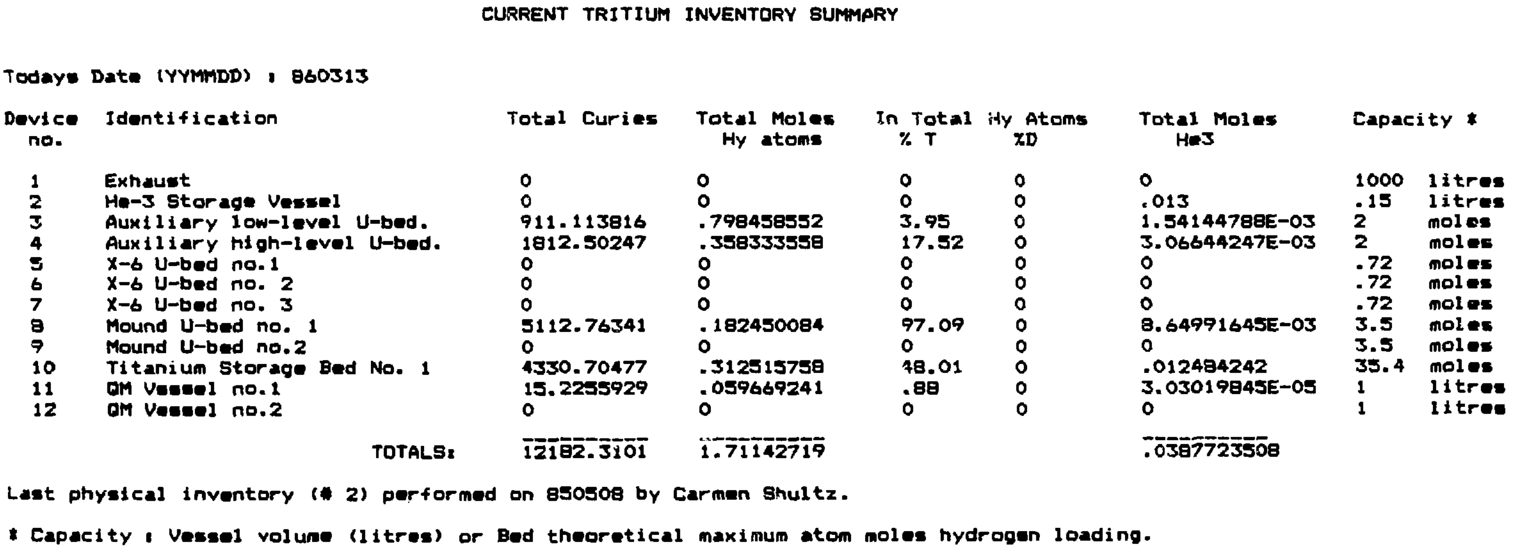
* 1 rem = 0.01 Sv



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                          CURRENT TRITIUM INVENTORY SUMMARY


Todays Date (YYMMDD) : 860313

Device  Identification                 Total Curies  Total Moles  In Total Hy Atoms  Total Moles     Capacity *
  no.                                                  Hy atoms      % T      %D       He3

   1    Exhaust                         0            0              0        0       0               1000  litres
   2    He-3 Storage Vessel             0            0              0        0       .013            .15   litres
   3    Auxiliary low-level U-bed.      911.113816   .798458552     3.95     0       1.54144788E-03  2     moles
   4    Auxiliary high-level U-bed.     1812.50247   .358333558     17.52    0       3.06644247E-03  2     moles
   5    X-6 U-bed no.1                  0            0              0        0       0               .72   moles
   6    X-6 U-bed no. 2                 0            0              0        0       0               .72   moles
   7    X-6 U-bed no. 3                 0            0              0        0       0               .72   moles
   8    Mound U-bed no. 1               5112.76341   .182450084     97.09    0       8.64991645E-03  3.5   moles
   9    Mound U-bed no.2                0            0              0        0       0               3.5   moles
   10   Titanium Storage Bed No. 1      4330.70477   .312515758     48.01    0       .012484242      35.4  moles
   11   QM Vessel no.1                  15.2255929   .059669241     .88      0       3.03019845E-05  1     litres
   12   QM Vessel no.2                  0            0              0        0       0               1     litres
                                        -----------  -----------                     -----------
                              TOTALS:   12182.3101   1.71142719                      .0387723508

Last physical inventory (# 2) performed on 850508 by Carmen Shultz.

* Capacity : Vessel volume (litres) or Bed theoretical maximum atom moles hydrogen loading.
```

FIGURE 16: OUTPUT OF CRNL TRITIUM LABORATORY ACCOUNTING PROGRAM

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5. ACKNOWLEDGMENT

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APPENDIX I:[*] Recovery and Packaging of Tritium from Canadian Heavy Water Reactors, W.J. Holtslander, T.E. Harrison, V. Goyette, and J.M. Miller.

ABSTRACT

The Tritium Extraction Plant being built at Chalk River Nuclear Laboratories (CRNL) will be the first industrial scale demonstration of the Liquid Phase Catalytic Exchange (LPCE) process for transfer of tritium from heavy water to deuterium. The plant will also demonstrate new technology in the areas of electrolytic cells for D_2 generation, water cooled recombiners, metal hydride packaging and magnetically coupled blowers for tritium service. It will be used to detritiate the heavy water in Atomic Energy of Canada Limited's (AECL) reactors.

INTRODUCTION

CANDU heavy water reactors produce tritium in the moderator and coolant circuits through neutron absorption by the deuterium atoms in heavy water. The concentration of tritium, in the form of DTO molecules builds up slowly with time of reactor operation. The mole ratio of DTO/D_2O after 12 years of operation of the Pickering reactors is about 2.0×10^{-5} (30 Ci/kg) in the moderator and less than 1×10^{-6} (2 Ci/kg) in the coolant. Eventually the tritium concentration will reach an equilibrium value where the production rate is balanced by the decay rate. This value is between 4 and 5×10^{-5} moles DTO/ mole D_2O (50-75 Ci/kg). Both Ontario Hydro and AECL are constructing plants to remove tritium from heavy water and to maintain the tritium concentration below the equilibrium value.

In addition to reducing reactor operator exposure to tritium, the AECL plant being built at CRNL will be used to demonstrate several new technologies on an industrial scale.

The tritium extraction process is made up of three parts. The first part or "front end" transfers the tritium and protium from the water molecule to a hydrogen molecule. In the heavy water case, this is represented by,

$$DTO + D_2 \overset{\text{catalyst}}{\rightleftharpoons} D_2O + DT$$

In the Chalk River plant the catalyst for this isotopic transfer will be the wetproofed catalyst (1,2) employed in the Liquid Phase Catalytic Exchange (LPCE) process (3,4). The plant will provide an industrial scale demonstration of the wetproofed catalyst for isotopic exchange. The Grenoble tritium removal plant, operating in France (5) and the Ontario Hydro Plant under construction at the Darlington Generating Station (6) both use vapour phase isotopic exchange. The second part of the process or "enrichment end" concentrates the tritium by low temperature distillation of the D_2/DT mixture to produce streams of essentially pure D_2 and T_2. The third section of the process is packaging of the pure T_2 in a safe way for long term storage or for commercial uses.

PROCESS DESCRIPTION

A schematic diagram of the process flowsheet is shown in Figure 1.

The LPCE column operates at 50°C and slightly above atmospheric pressure. Heavy water trickles down the column while deuterium gas flows up the column. The heavy water stream is purified and degassed before it enters the LPCE column and purified again as it leaves. The extraction of tritium and protium from heavy water in a single pass is expected to average 80 and 90 percent, respectively.

The LPCE column is 0.15 m in diameter and has five sections with a liquid distributor in each. The bottom section is a humidifier for deuterium gas and is filled with inert packing. The four upper sections are filled with wetproofed catalyst. This catalyst is available in both random and ordered bed forms (7) and these forms will be demonstrated one at a time in the LPCE unit.

Provisions are made for reactivating the catalyst in situ by passing air at 150°C through it, when required.

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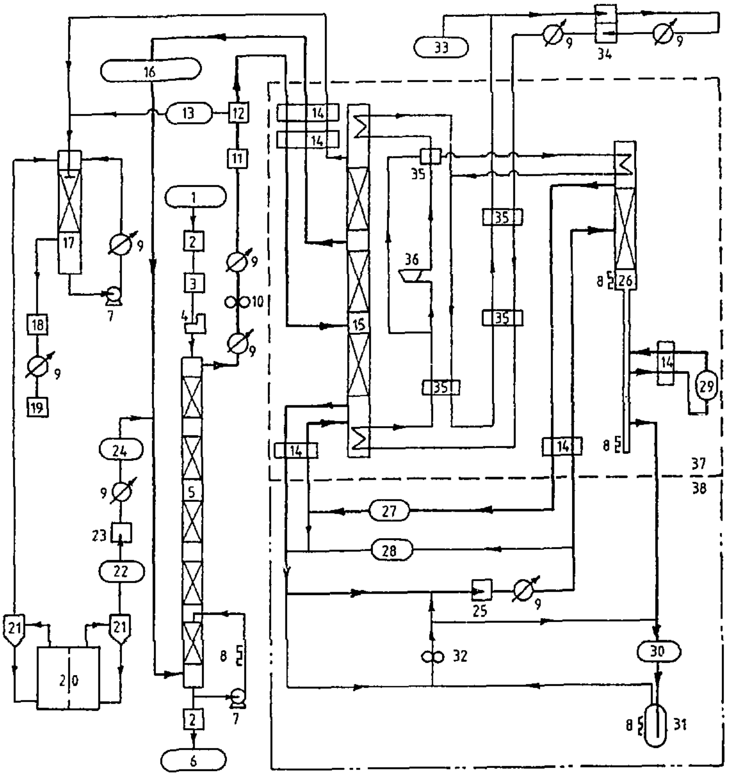

1. D_2O Feed Tank
2. Disposable Purifier
3. D_2O Degasser
4. Metering Pump
5. LPCE Column
6. D_2O Product Tank
7. D_2O Pump
8. Electric Heater
9. Cooler
10. Deuterium Blowers
11. Freezer-Dryers
12. Air Adsorbers
13. Off-gas Tank
14. D_2 Heat Interchanger
15. Distillation Column #1
16. D_2 Expansion Tank
17. D_2/O_2 Recombiner
18. Backup Recombiner
19. Off-gas Dryer
20. Electrolytic Cell
21. Separator/Demister
22. D_2 Surge Tank
23. D_2 Makeup Compressor
24. D_2 Makeup Tank
25. DT Circulator
26. Distillation Column #2
27. DT Expansion Tank
28. DT Storage Tank
29. DT/T_2 Converter
30. T_2 Metering Tank
31. T_2 Package
32. Moving-Spiral Pump
33. He Storage Tank
34. Helium Compressor
35. He Heat Interchanger
36. He Turbo-expander
37. Cold Box
38. N_2 Purged Glovebox

Figure 1: Schematic Diagram of Process Flowsheet

Deuterium gas from the LPCE column is dried, purified and delivered to a cryogenic distillation system where tritium and protium are concentrated and separated from the deuterium stream.

Two magnetic drive vane blowers working in parallel are used to circulate deuterium gas. These blowers are manufactured by Nova Magnetics Limited of Dartmouth, Nova Scotia. This application represents the first time such blowers have been used in tritium service. Some of the moisture in the deuterium stream is removed by water cooled condensers on the inlet and outlet of the deuterium blowers. The remaining moisture is removed by alternating use of one of two freezer-dryer units cooled by liquid nitrogen.

Gas impurities are removed from the deuterium stream by one of two air adsorbers that are filled with molecular sieve and cooled by liquid nitrogen. These units are switched over and regenerated about once per month. During regeneration at 300°C under vacuum, the desorbed deuterium is led off through an off-gas tank to a wetproofed catalyst recombiner.

After passing through the cryogenic distillation unit the deuterium stream returns to the LPCE column via an expansion tank. This tank limits the pressure in the system to about 500 kPa absolute when the liquid deuterium inventory in the first of two distillation columns boils off during plant shutdowns.

Tritium and protium are concentrated and separated from deuterium by fractional distillation in two columns operating at about 25 K. The cryogenic equipment is being designed and manufactured in Switzerland and supplied through Sulzer Canada Inc.

The two columns are located inside a 15 m high cold box. All gas streams enter and leave the cold box through heat interchangers to minimize the refrigeration requirements. The first distillation column is 0.2 m in diameter and is filled with Sulzer CY packing. The purified deuterium stream passes upwards through the middle portion of this column and then returns to the LPCE unit. In the upper portion of the column, protium is stripped out, and bled off continuously to the recombiner. In the lower portion of the column, tritium is enriched by a factor of 25 above its concentration in the deuterium feed stream. This relatively low factor was chosen to minimize the tritium inventory in the major deuterium (D_2) circuit of the plant where there is no secondary containment.

The second distillation column is made up of three sections. The diameter of the upper section is 50 mm and the two lower sections, each 12 mm. A stream of the tritium-enriched deuterium gas is delivered from the first distillation column to the middle of the upper section of the second column while a tritium-depleted stream returns from the top of the second distillation column to the bottom of the first. Tritium is enriched in the remainder of the second distillation column.

A catalytic converter to generate D_2 and T_2 from DT is located in a line between the two lower sections of the column. Nearly pure tritium accumulates in the bottom of the second column and is bled off in small batches to the packaging system.

During plant upsets, the two distillation columns are isolated from each other and the liquid inventory in the second column boils off into an expansion tank. A duplicate of this tank is provided for maintenance purposes. The diaphragm compressor and moving-spiral metal bellows vacuum pump can be used in tandem to pump the contents of the second distillation column into the storage tank. The metal bellows pump is used to recirculate gas through the tritium packaging system when necessary.

Any tritium that leaks from the process equipment will be released to atmosphere. For safety reasons, equipment containing high concentrations of tritium is located either in the cold box or in a nitrogen purged glovebox.

Off-Gas System

The off-gas system is designed to accommodate a continuous stream of protium-rich gas from the first distillation column, the deuterium produced by the monthly regeneration of the air adsorber, and the total inventory of deuterium whenever the low tritium section of the plant is evacuated for maintenance. All of these sources of deuterium will be recombined to form water in a water-cooled recombiner using wetproofed catalyst. For this application the recombiner is 0.15 m in diameter, and can handle 0.1 to 3 std m^3/h. A larger version of this type of recombiner has been purchased by the US Department of Energy for the Idaho Falls Laboratory.

Deuterium is fed to the recombiner through a distributor located in the catalyst bed. Cooling water and a slight excess of oxygen are fed at the top of the column. All streams flow down through the bed. The water warms up to about 80°C in passing through the bed, and most of this water is cooled to 30°C and recirculated. The excess oxygen left after recombination has taken place is vented through a small backup recombiner to ensure recombination is complete, and then through a cooler and dryer to recover heavy water.

Makeup System

All of the deuterium that has been dumped to the off-gas system must be replaced, and at a similar rate. An electrolytic cell with an output of 0.1 to 3 std m^3/h of deuterium gas has been selected. The cell is a prototype of a low water inventory electrolytic cell developed for tritium service under an Ontario Hydro-AECL contract to Electrolyser Inc. (co-owned by the Electrolyser Corporation Limited and Noranda Mines Limited).

The prototype unit is a low pressure single cell with an asbestos diaphragm, KOD electrolyte, external gas separators, coolers and demisters, and a heavy water inventory of only 3 litre/kiloampere.

Pressure controls are provided so that there is no differential pressure across the asbestos diaphragm. The oxygen produced will be fed directly to the off-gas recombiner. The deuterium will flow to a surge tank and will then be compressed by a diaphragm compressor to 500 kPa pressure and stored in a tank from which it will be fed to the deuterium circuit as needed.

Refrigeration System

A helium gas refrigeration circuit provides refrigeration at 20 K to the cryogenic distillation system. A two-stage reciprocating compressor compresses the helium from 300 to 1300 kPa absolute, and the heat of compression is removed by water-cooled heat exchangers. The helium flows to the cold box where it passes through heat interchangers and then, at 30 K, provides heat to the reboiler of the first distillation column. After further heat exchange the helium is expanded through a gas-bearing turboexpander, where its temperature drops to 15 K. The gas then passes through the condenser of the first distillation column and returns to the compressor via the heat interchangers that cool the compressed helium. A helium sidestream provides the condensing capacity in the second distillation column, while reboil in this column is provided electrically.

MODELLING OF LPCE AND CRYOGENIC DISILLATION COLUMNS

Process model simulations of the LPCE (8) and cryogenic distillation columns have been written to provide design information and to study operating parameters.

The model of the LPCE column is based on the two reactions:

$$DTO_l + D_2O \rightleftharpoons DTO_v + D_2O_l$$
$$DTO_v + D_2 \rightleftharpoons DT + D_2O_v$$

Where the subscripts l and v refer to liquid and vapour.

The mass transfer rate equations (9) are given by:

$$\frac{dY}{dZ} = \frac{-\rho_{KR}}{G}\left[\alpha_R Y(1-\nu) - \nu(1-Y)\right]$$

$$\frac{dX}{dZ} = \frac{-\rho_{KD}}{L}\left[\alpha_D \nu(1-X) - X(1-\nu)\right]$$

where ρ_{KR}, ρ_{KD} in moles/m^3.s are the catalytic and vapour-liquid overall rate constants and α_R, α_D are separation factors. Y, X and ν refer to concentration in the gas (D_2), liquid (D_2O_l) and vapour (D_2O_v) phase respectively, Z is the column length, G is the gas flow rate and L is the liquid flow rate.

On the basis of constant flux of isotope through any infinitesimal section of the column the vapour concentration can be eliminated from the rate equations. The program then integrates upwards from the bottom of the column using a Runge-Kutta integration routine.

A computer code named "CRYGEN" was produced to study the steady-state operation of two cryogenic distillation columns and a catalytic converter.

The inputs required by the simulation are: internal liquid and gas flows, pressure profile, bottom concentrations for column 1 and 2, first column top plate HD concentration, transfer flow between columns, equilibrium constant for the converter, equilibrium adjustment factors and the total number of plates in each column.

The model incorporates differences in latent heat of vaporization among the isotopic species and nonideality of the hydrogen isotope solutions. The iterative procedure calculates the temperature, flows and composition on each plate.

The catalytic converter is simulated by solving the equilibrium and mass balance equations using an iterative procedure.

PACKAGING OF THE TRITIUM PRODUCT

It is intended to package the T_2 product in a form that allows safe storage indefinitely. To meet this requirement the tritium had to be immobilized so that in event of failure of the container, tritium would not be released. This requirement eliminated gas storage. It also ruled out storage as a uranium tritide because of its pyrophoric property.

Metal hydrides appeared to offer the best promise to meet these requirements (8). In addition to being able to bind the tritium strongly, it was also desirable to find a metal where the hydride could be formed in a simple reaction, preferably at room temperature. While packaging for safe storage was the primary objective, it was also recognized that because a commercial market for tritium exists it should be possible to recover the tritium.

Work at CRNL has centred on the immobilization of tritium as titanium and zirconium tritides. The hydrides of both of these metals fit well with the requirements. Examination of the methods to prepare the hydride have shown (10,11) they can be formed readily by bringing together the activated metal and pure T_2 at room temperature. The metal, in sponge form, is activated by heating to 400-450°C in vacuum for approximately one hour. This is believed to dissolve the surface oxide into the bulk of the metal. The hydrogen must be pure, particularly free of oxygen and moisture. In the tritium extraction plant this is the case, since the gas is received from the low temperature distillation column. In the case of tritium, there is always some ^{3}He from decay and special provisions must be taken to accommodate it.

Typical hydriding with titanium sponge occurs quickly at the initial temperature of 25°C. However, with even less than 1% helium in the gas the hydriding reaction is stopped by the blanketing effect of the helium. Provision of a circulator in the packaging system to move the gas through the metal bed overcomes this problem.

The stability of these hydrides in air and water has been studied. The leach tests of titanium tritide have been reported elsewhere (12). The test results show a very low release rate of tritium into water from titanium tritide. Cumulative fractional release rates of (0.3-1) x 10^{-4} have been observed over a period of 778 days. These tests have been extended to samples containing high specific activity tritium. Cumulative fractional releases from these samples was in the range of (1-2) x 10^{-5} for $TiT_{1.8}$ over 150 days.

The hydride or tritide of titanium is brittle and there is a tendency of the sponge material to become finer as it undergoes physical shock. To know if tritium would be released from exposure of the fines to air in event of failure of the container, it was important to determine the ignition temperature of titanium and titanium hydride particles. The ignition temperature has been measured as a function of particle size. As expected, lower ignition temperatures are observed for the smaller particle size range. They range from 660°C for the starting material at 4-6 mm to 420°C for the finest particle size range <38 µm.

It is possible to have ignition of the titanium hydride at room temperature if fine enough particles are present. However, tests with titanium hydrides that have undergone 10 hydriding-dehydriding cycles and physical agitation for a number of weeks show less than 10% of the material in the <38 µm range. Even this fraction has an ignition temperature much above the temperature expected in storage locations.

The tritium for the CRNL plant will be immobilized in and stored in a 6 L stainless steel container shown schematically in Fig. 2 with its transportation package. The primary storage container has 850 g of Ti sponge in it which when loaded to an average of $TiT_{1.0}$ can package 50 g (0.5 MCi) per vessel. The vessel is designed to contain the ^3He from complete decay of the tritium. For transportation the primary vessel will be placed in a secondary container and then in an insulated drum. The vessel acts both as the reactor for the hydriding reaction as well as the primary container. Unpublished work at the CRNL tritium laboratory has demonstrated the immobilization of concentrated tritium in a vessel of this design.

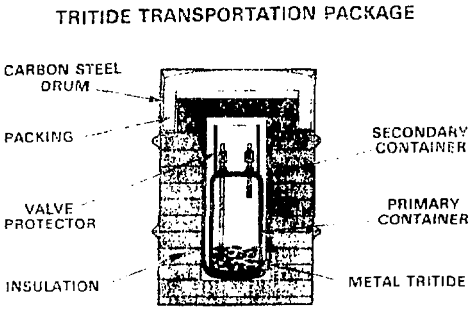

Figure 2: TRITIDE TRANSPORTATION PACKAGE

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APPENDIX 2:* Tritium Instrumentation for the Chalk River Nuclear Laboratories Tritium Extraction Plant, E.C. Davey, R.G.C. McElroy, and S. Kupca.

ABSTRACT

A Tritium Extraction Plant (TEP) is under construction at the Chalk River Nuclear Laboratories (CRNL) for the extraction of tritium from the heavy water moderator and coolant of research and power reactors owned by Atomic Energy of Canada Limited. Since the process equipment will contain a large inventory of tritium (~20 g), plant operating personnel must be continually aware of process stream activity levels for economic reasons and promptly alerted in the event of leakage for personnel health protection purposes and inventory loss minimization. Consequently, the TEP is equipped with a number of tritium monitors for the continuous measurement of tritium contained in the process equipment, the building air and the plant exhaust stacks. This paper outlines the approach taken to provide tritium monitoring at the TEP for health protection, environmental and process requirements and describes the general features of specific monitors in each classification.

BACKGROUND

The Tritium Extraction Plant (TEP), presently under construction at the Chalk River Nuclear Laboratories (CRNL), will extract tritium from the heavy water moderator and coolant of research and power reactors owned by Atomic Energy of Canada Limited. In spite of an excellent record in engineering systems to minimize moderator and coolant losses, the chronic leakage of tritiated heavy water of concentration up to 740 GBq/kg (~20 Ci/kg) from the reactor systems has become responsible for a significant fraction (~15%) of the radiation exposure of some operating personnel. The contribution of tritium to the effective dose equivalent of these personnel would be expected to increase as the tritium concentration in the heavy water continues to build up to its equilibrium concentration of about 1.5 TBq/kg (~40 Ci/kg). Periodic removal of tritium from the moderator and coolant of these reactors is expected to yield a significant reduction in the average effective dose equivalent from tritium for operating personnel and the tritium emissions to the environment from these facilities.

The TEP consists of two buildings linked by an interconnecting corridor as shown in Figure 1. Tritium extraction takes place in the process building, which contains the liquid phase catalytic exchange columns, cryogenic distillation system and tritium handling gloveboxes. Support facilities for the extraction process, such as the control room, cryogenic refrigeration equipment, maintenance shop and feed and product storage areas, are housed in the service building. During routine operation, only the service building will be occupied on a continuous basis.

The TEP has been designed to approximately 80% of the tritium per processing pass. The the annual extraction capacity will be about 50 g elemental tritium with a 185 GBq/kg (~5 Ci/kg) feed concentration. Tritium extraction will take place in two processing steps. First, tritium from the heavy water feed is transferred to a deuterium gas stream in the presence of a CRNL developed wet-proof catalyst. Second, pure tritium gas is removed from the deuterium gas by fractional distillation at 25 K. During operation of the extraction process, approximtely 7.4 PBq (~200 kCi) of tritium will

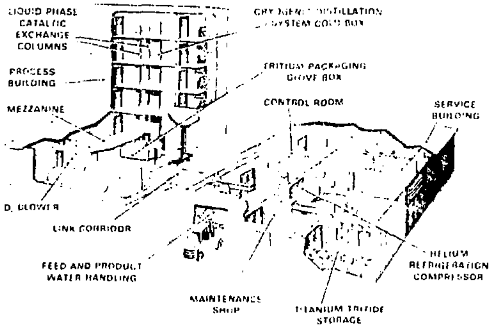

Figure 1: CRNL TRITIUM EXTRACTION PLANT

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be present within the process equipment. The extracted tritium will be stored as a metal hydride in sealed containers, each with a capacity for 50 g of tritium.[1]

The derived release limit (DRL) of tritium (HTO) for the TEP is determined by the onsite exposure to CRNL personnel. This limit is 78 TBq/week (2.1 kCi/week). Although tritium can be released from the plant in either the gaseous (HT, DT, T_2) or the more hazardous oxide (HTO, DTO, T_2O) form, the safety analysis and DRL are based on the conservative assumption that any tritium released is in the oxide form.

The exposure of operating personnel to tritium releases is minimized by three design features. Firstly, subsystems with the potential for large tritium releases are surrounded by secondary enclosures. For example, the tritium packaging equipment is contained within a sealed and nitrogen purged glovebox. Secondly, both buildings have a non-recirculating airflow with 10 air changes per hour. Thirdly, the TEP is equipped with a number of monitors for the continuous measurement of the tritium present in the building air, the plant exhaust stacks and the process streams. These tritium monitors can be classified according to function: health protection, environmental and process.

HEALTH PROTECTION MONITORS

The primary purpose of the health protection monitors is to measure tritium-in-air concentrations so that operating personnel can adequately estimate the radiological hazard to workers in a given area. These monitors also serve a secondary purpose in alerting operating personnel to failures of the process containment or equipment. Such failures may first appear as increases in tritium activity in the room air which can be readily detected by the facility health protection monitors.

All the health protection monitors consist of ion chamber-based instruments. Three types of monitors are in use:

- tritium area monitors to measure total tritium-in-air concentrations of selected areas,
- portable tritium monitors for manual surveys of unmonitored areas for both total tritium and HTO contributions, and
- a discriminating tritium monitor for remote characterization of total tritium or HTO hazards within several areas of the plant.

The characteristics of each of the three types of monitors are outlined in Table 1.

Eight tritium area monitors are used to continuously monitor the plant air in specific locations and annunciate the presence of abnormal tritium-in-air concentrations. All of these monitors measure total tritium; they are not species specific. Thus, all readings from these monitors are interpreted with the conservative assumption that the tritium measured is in the oxide form. Consequently, the radiological hazard to personnel will be overestimated for HT leakage.

The locations to be monitored by the area monitors are as follows:

Process Building

- the north side of the mezzanine,
- the south side of the mezzanine,
- the top of the D_2 blower,
- the top of the tritium packaging glove box,
- the tritium packaging glove box exhaust,
- the cryogenic distillation cold box pump discharge,

Service Building

- the feed water and product handling area, and
- the maintenance shop.

All of these monitors are based on a CRNL-designed ion chamber/electrometer detection unit manufactured by Scintrex Limited of Concord, Ontario. The electrometer in this unit is capable of auto-ranging operation spanning 7 decades. However, for the TEP area monitor application only a single four decade range from 0.037 MBq/m^3 (1 $\mu Ci/m^3$) to 370 MBq/m^3 (10^4 $\mu Ci/m^3$) is utilized. Each monitor is equipped with a local digital display for direct indication of tritium activity level by nearby personnel. A 4-20 mA output is provided for the connection of remote displays.

For the monitors mounted inside the process building, the potential hazard from explosive mixtures of hydrogen gas and air dictated that the monitors be mounted in an explosion proof environment. To meet this requirement, each monitor is mounted in a ventilated enclosure. Each enclosure contains the detection unit, local power supplies, digital display and sampling pump for one monitor.

To supplement the fixed area monitors, plant operations personnel are equipped with two hand carried portable tritium monitors, permitting the monitoring of air for tritium in areas remote from the sampling points of the area monitors. The portability of these units enables personnel to sniff out the location of small leaks for repair.

Type	Number Deployed	Measurement Technique	Range (MBq/m^3)	Resolution (MBq/m^3)	Response Time (min)
Fixed Area	8	Ion chamber	0.037 to 370	0.074	< 1
Portable	1	Ion chamber	0.37 to 740	0.4	< 1
	1	Ion chamber with desiccant	0.37 to 740	-	-
Discriminating	1	Ion chamber and Nafion separator	0.037 to $3.7x10^5$	0.074	< 1 (Total Tritium Channel) < 2 (HTO Channel)

Note: 1 Mbq/m^3 is equivalent to 27 $\mu Ci/m^3$.

TABLE 1 - Health Protection Tritium Monitors

This portable monitor employs ion chambers of 80 cm^3 active volume and spans a measurement range of three decades from 0.37 MBq/m^3 (10 $\mu Ci/m^3$) to 740 MBq/m^3(2000 $\mu Ci/m^3$. The monitor was originally developed at CRNL[2] and is now manufactured under license by Scintrex Limited as the Model 209. Normally, the portable monitor measures total tritium-in-air concentrations. For the TEP application, it was judged to be useful to have one of the two monitors with a limited capability to differentiate between the gaseous and oxide tritium species. Consequently, one monitor is currently being outfitted with a manually selectable sampling system based upon a desiccant to achieve the preferred response.

The third type of health protection monitor is the discriminating tritium monitor. This monitor is capable of measuring simultaneously the total tritium and tritium oxide concentrations of an air sample. For the TEP application, the monitor is outfitted with a manual sampling manifold for manual selection of one of up to 25 sample lines from around the plant. The sample points consist of the air samples from each of the area monitors, the process building stack monitor and sampling points associated with relief valves of the process equipment. In this configuration, the monitor serves three separate roles.

Firstly, the monitor serves as a diagnostic instrument to supplement the array of non-discriminating area monitors. The eight area monitors, set to alarm on the conservative HTO assumption, provide a prompt local assessment and alarm of total tritium activity. The discriminating monitor provides a more precise assessment of the radiological tritium hazard in a given area through the separate measurement of total tritium and HTO concentrations. Thus, once an abnormal concentration of tritium is identified in a given area by the local area monitors, the discriminating monitor is used to provide a more precise assessment of the hazard by quantifying the concentration of each tritium species.

Secondly, the monitor serves in a diagnostic role in locating and characterizing the nature of tritium releases via any of the process relief valves. The first indication of such releases will normally be indicated by the process building stack monitor. Once an increase in stack tritium concentration above chronic background levels is detected, operating personnel will use the discriminating monitor to first localize the release to a given relief valve and then quantify the release as to species type.

Thirdly, the monitor serves as a spare for any of the eight area monitors or the process building stack monitor in the event of a monitor failure.

The monitor for the TEP has evolved from the discriminating monitor development work of McElroy, et al.[3] It is implemented from two basic modules, a Nafion (DuPont) membrane separation unit and a tritium area monitor currently under development at Scintrex Limited to AECL specifications.

The separation module consists of a Nafion membrane assembly and three process control loops to maintain sample flows, humidity and temperature within acceptable operating limits. The area monitor module consists of a detection unit identical to those used for the eight area monitors and a display and control unit which provides signal-to-noise enhancement, alarm annunciation, local activity display and remote communications functions. For the extraction plant application, the discriminating monitor will be deployed with two area monitors and a single sample separation module. Such a configuration permits the total tritium and HTO contributions of a given air sample to be measured simultaneously. The monitor is capable of measuring airborne tritium concentrations in either channel in the range from 0.037 MBq/m^3 (1 $\mu Ci/m^3$) to $3.7x10^5$ MBq/m^3 (10^7 $\mu Ci/m^3$) with a resolution of 0.074 MBq/m^3 (2 $\mu Ci/m^3$) in the lowest range.

In addition to the dedicated monitoring facilities previously described, the operating personnel have access to two other means of health protection monitoring from alternative facilities at CRNL. Conventional sampling bubblers are available to characterize tritium-in-air concentrations which are below the lower range limit of the area monitors. To monitor for surface tritium contamination, liquid scintillation counting of swipe samples is available.

ENVIRONMENTAL MONITORS

The TEP is not equipped with a tritium cleanup system. Tritium escaping from the process is diluted in the plant air and promptly exhausted via one of the two building ventilation stacks. Any release of tritium to the surrounding environment is measured by monitoring the stack tritium releases.

Two forms of environmental tritium monitoring are employed for this purpose. Compliance monitoring is used to provide a continuous measurement of the environmental tritium release to demonstrate that the plant tritium releases are within the regulatory release limits defined by the operating license. Control monitoring is used to provide timely detection and annunciation of increases in environmental tritium release where the continued tritium emission would result in administrative and/or regulatory release limits being exceeded. The three monitors selected to supply the environmental tritium monitoring functions are listed in Table 2.

For compliance monitoring purposes, each building stack is equipped with a sampling bubbler to measure the integrated tritium release in a given time period. To ensure that gaseous tritium releases are properly accounted for, each bubbler is equipped with a sample conditioning front end which converts any gaseous tritium to the oxide form in the presence of a CRNL-developed catalyst. At the end of each sampling period, a sample of the bubbler contents is assayed by liquid scintillation counting to determine the environmental tritium release from each stack. During normal plant operation, this measurement technique enables plant personnel to characterize chronic tritium releases to below 0.01% of the DRL.

To simplify manual sampling and maintenance procedures, both bubblers are installed remotely from the stacks in the corridor connecting the service and process buildings and are connected to the stacks via sampling lines.

Since the potential for significant release of tritium from within the service building on a routine basis is very small, only the process building is equipped with a control monitor. The process building stack monitor consists of an ion chamber/electrometer detection unit identical to those deployed as area monitors for health protection purposes. To assure a short response time to changes in stack activity, the monitor is placed adjacent to the stack base in the upper floor of the tower portion of the process building. Since many relief valves for the extraction process are vented only to the upper floor of the process building tower, the stack monitor will provide the first indication of tritium releases from the process via the relief valves.

As with the other fixed area monitors in the process building, this unit is housed inside a ventilated enclosure to prevent the hydrogen/air concentration adjacent to the monitor from reaching explosive levels.

The nominal flowrate in the process building stack is 4.3 m^3/s. Since the DRL for the facility is 78 TBq/week (2100 Ci/week), a chronic building tritium air concentration of 0.3 MBq/m^3 (8.2 $\mu Ci/m^3$) is sufficient to result in an environmental release of 1% of the DRL. The monitor has an inherent sensitivity of 0.074 MBq/m^3 (2 $\mu Ci/m^3$) and is thus capable of resolving 0.25% of the DRL.

Sampling Point	Purpose	Measurement Technique	Range (MBq/m^3)	Resolution (MBq/m^3)	Response Time (min)
Service building stack	Compliance monitoring	Sampling bubbler and liquid scintillation counting	<0.003 to $>3\times10^2$	<0.003	-
Process building stack	Compliance monitoring	Sampling bubbler and liquid scintillation counting	<0.003 to $>3\times10^2$	<0.003	-
Process building stack	Control monitoring	Ion chamber	0.037 to 370	0.074	1

Note: 1 MBq/m^3 is equivalent to 27 $\mu Ci/m^3$.

TABLE 2 - Environmental Tritium Monitors

PROCESS MONITORS

The species of interest in monitoring the extraction and storage of tritium is gaseous tritium (HT,DT,T_2). Five ion chamber based monitors, being developed at CRNL, will be used for the measurement of tritium gas concentrations associated with the cryogenic distillation and tritium packaging processes. The locations of these monitors are shown on the process flowsheet for the plant (Figure 2).

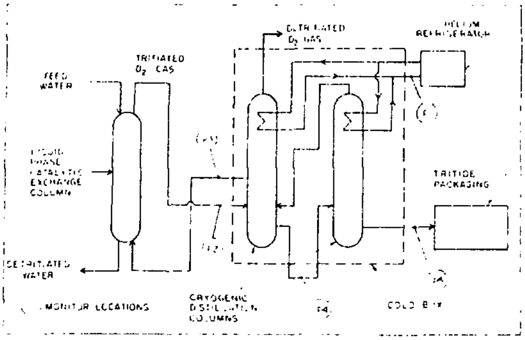

Figure 2: TEP PROCESS FLOWSHEET AND MONITOR LOCATIONS

Each monitor is situated on a bypass sampling line associated with a specific aspect of the extraction process. One monitor, P1, is used to monitor for leaks from the cryogenic distillation column feed to the helium refrigerant via the heat exchangers. The four remaining monitors, P2 through P5, are used to monitor the feed, tails and product tritium concentrations associated with the two cryogenic distillation columns. The information from these monitors will assist operations and design personnel to assess the extraction efficiency of each cryogenic distillation stage. The measurement characteristics of each of these monitors are detailed in Table 3.

The tritium activity signals from these monitors are not used to directly control process parameters. Instead, plant operators will be responsible for monitoring all process monitor readings and initiating any corrective action necessary according to established control procedures.

In developing these monitors, the designers were faced with four specific challenges as imposed by the application. These challenges were:

- to implement the monitors with standard components to minimize customizing costs,
- to accommodate sample pressures of up to 2 MPa,
- to minimize the signal background due to adsorption of tritium on surfaces within the detector, and
- to qualify the monitors for operation in potentially explosive environments.

In meeting these challenges, the following approaches were taken. A standard detection geometry was adopted for four out of five monitors and measurement ranges and sensitivities for these four monitors were customized

	Sampling Point	Purpose	Measurement Technique	Range (TBq/m^3)	Resolution (TBq/m^3)	Response Time (min)
P1	Inlet to helium compressor	Process monitoring	Ion chamber	0.0004 to 0.004	0.0004	~1
P2	Feed to first distillation column	Process monitoring	Ion chamber	0.007 to 7	0.007	~1
P3	Tails product of first distillation column	Process monitoring	Ion chamber	0.004 to 0.4	0.004	~1
P4	Feed to second distillation column	Process monitoring	Ion chamber	0.2 to 200	0.2	~1
P5	Metering tank (product of second distillation column)	Product monitoring	Ion chamber	90 to 90,000	90	~1

Note: 1 TBq/m^3 is equivalent to 27 μCi/m^3.

TABLE 3 - Process Tritium Monitors

by electronic component substitution. This common detection unit geometry, based on an ion chamber with 300 cm^3 active volume, is used for all monitors except P5. For the higher range product monitor, P5, an ion chamber with only 15 cm^3 active volume is used Wherever possible, pre-qualified commercial components have been used to minimize custom machining and in-house qualification testing. For example, in meeting the pressure requirement, standard pipe fittings conforming to ANSI standards were used to construct a pressure vessel enclosure. Within this pressure boundary, the flow-through ion chamber and sample routing baffles are mounted, thus removing the need to build an ion chamber to satisfy the pressure requirements explicitly.

Previous experience at CRNL with the use of an ion chamber of 3 cm^3 active volume to monitor the production of tritium for radioisotope purposes had shown that the background signal from the adsorption of tritium on surfaces within an ion chamber can become very large, especially when the ion chamber is exposed to high activity tritium gases. In minimizing this potential problem, two aspects of the ion chamber design are worth noting. Firstly, the active volume of the ion chambers was increased to boost the signal portion of the ion chamber current. Secondly, the surface of the active volume was minimized by using a central axial wire collecting electrode surrounded by a wire mesh for the high voltage electrode. These two design changes are expected to increase the signal to background ratio by a factor of 20 over that of the earlier design.

Finally, to avoid complicating the monitor design with an inherent explosion proof capability, a separate enclosure purged with nitrogen gas will be used to house all monitors. Such an approach was judged to be the most cost effective for the extraction plant application, since several other instruments are being mounted within the enclosure to satisfy explosion proof require ments as well.

Each of the five monitors is equipped with an on-board electronic package comprising an electrometer, log converter, 4-20 mA driver and a local display.

DATA TRANSMISSION, ARCHIVING AND ALARM ANNUNCIATION

All monitors except for the portable health protection units and the stack bubblers will have their outputs transmitted to a central console in the service building control room. The signal output of each monitor is converted to a log format and then transmitted via the 4-20 mA signal standard to the control room signal receivers. This method of signal conversion and transmission permits a signal range of up to four decades which is adequte for the TEP application.

In the control room, the received signals will be used for three purposes. Firstly each monitor output will drive a dedicated display unit, calculated in units of $\mu Ci/m^3$. Secondly, each monitor is connected to a dedicated alarm unit. Each alarm unit has a manually adjustable alarm setpoint. Thus, annunciation of local alarms associated with each monitor in the plant is controlled via alarm logic signals transmitted from the control room to each monitor field location. Thirdly, each monitor output is routed to a common data logger for the continuous recording of the tritium activity level in each monitored location.

In addition to the local and control room displays for the process building area monitors, an additional set of tritium activity displays is situated in the corridor linking the process and service buildings. These displays allow operations personnel to review process building tritium activity levels immediately prior to entry.

COMMISSIONING STATUS

The complement of monitors discussed in this paper are currently undergoing engineering development at CRNL or are being procured through commercial instrumentation suppliers. All units are scheduled to be available for installation and commissioning at the TEP in 1986 January.

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APPENDIX 3:* The Licensing Aspects of the Tritium Extraction Plant at the Chalk River Nuclear Laboratories, P.R. Ballantyne.

ABSTRACT

The Tritium Extraction Plant is being built at the Chalk River Nuclear Laboratories and requires licensing to be operated. Some aspects of this licensing are discussed including the licensing authority and the general licensing procedure of a new facility. Since dose hazard is an important part of the licensing, various aspects of estimating possible doses are discussed. These aspects played a major role in establishing the code to which the plant is being designed and built.

INTRODUCTION

The Tritium Extraction Plant (TEP) is being built at Chalk River Nuclear Laboratories (CRNL) to reduce the tritium concentration in Atomic Energy of Canada (AECL) heavy water reactors, thus reducing the tritium contribution to whole body dose and the tritium emissions to the environment. The plant will also be used to demonstrate and gain experience in the operation of a tritium extraction plant and in particular the application of the wet proof catalyst in the catalytic exchange column and cold recombiner (Figure 1).

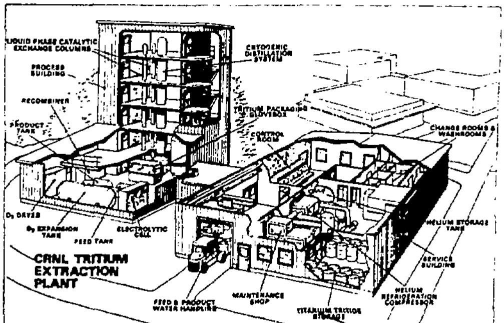

FIGURE 1 CRNL TRITIUM EXTRACTION PLANT

LICENSING OF AECL NUCLEAR FACILITIES

From the beginning of its operation, AECL ha been responsible for the safety review an approval of its own facilities. A safety revie and approval committee, the Nuclear Safet Advisory Committee (NSAC), with representative from across AECL as well as from the Atomic Energy Control Board (AECB), has acted as the regulatory body. It has been responsible for the safety review and approval of facilities at AECL sites, of which CRNL is one. In 1980, the AECB issued site licences to AECL and appointed a project officer to inspect the nuclear facilities, at these sites, for compliance with the terms of the site licences. The NSAC continues to conduct the detailed safety review of these facilities on behalf of the President of AECL and is responsible for reviewing and approving the design, construction, and operation of nuclear facilities at AECL's Research Company. Once a facility is reviewed and approved, a request is made to the AECB to amend the site licence to include the new facility. After the amendment is granted, the owner is authorized to operate the facility according to a set of "Principles and General Rules".

An NSAC document, AECL-MISC-191, contains a list of the nuclear facilities reviewed by the NSAC and licensed by the AECB, a set of principles and general rules for the operation of each of the facilities and a set of guideline documents. The guideline documents outline the criteria for deciding whether a facility requires a review by the NSAC and the procedures to be followed if such a review is deemed necessary.

Elements of Approval Process

The review and approval process, for a facility, involves up to five iterative steps of presentation, study and discussion involving the owner/operator, the committee and various consulting groups. These stages are: a letter of notification; concept review and location approval; construction approval; licensing, startup and commissioning; and submission of an updated FSAR.

Major document submissions include a Concep Safety Assessment Report (CSAR) requesting approval in principle of the concepts involved and the location of the facility, a Preliminary Safety Analysis Report (PSAR) requesting construction approval and a Final Safety Analysis Report (FSAR) requesting approval to startup, commission and operate the facility. The final submission requires an updated FSAR with relevant commissioning experience and

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becomes part of the permanent licensing document for the facility.

The safety reports for a facility deal with the safety of normal operations as well as abnormal situations. Potential accidents must be addressed and be demonstrated to have an acceptably low probability of occurrence and/or have acceptably low consequences. As part of the licensing program, an annual safety report must be produced for each licensed facility, for NSAC and AECB review. In addition, any "unusual occurrence" (as defined in the "Principles and General Rules" for the facility) or any significant changes or departures from the final safety analysis report, must be reported promptly to the Technical Secretary of the NSAC and to the AECB Project Officer.

Construction and Code Approval

To obtain construction approval, the NSAC reviews the PSAR of the proposed facility, calling upon specialists from within AECL, the AECB or elsewhere, as required, to assist with the review. The code, to which the process systems must be designed, is approved by the AECB. The code classification is obtained by formal submission of documents to the AECB and is based on the Canadian Nuclear Standard CSA Standard CAN3-N285.0-M81 [1].

Although the N285 standard was written primarily for application to CANDU Nuclear Reactors, it is being applied to other nuclear facility equipment. The evaluation is based on the estimated doses that various groups of people would receive if the system or component, under consideration, were to fail and cause the release of contained radioactive material.

For example, if a tank, containing some radioactive inventory, was to fail and result in an estimated dose of 10 mrem to the public and 12 rem to a plant operator, the tank would be designated as Class 2 (Table 1). The code class designations of 1, 2 and 3 correspond to the ASME Nuclear codes, Section III Class 1, 2, and 3 while the code class 6 designation corresponds to Section VIII.

RADIOLOGICAL HAZARD

To proceed with the design and construction and ultimately the licensing of the TEP, the radiological hazard must be evaluated. This permits the code, to which the process system should be designed, to be established. The safety philosophy is to design the TEP so that there is virtually no leakage of hydrogen isotopes out of the system. In the event of a hydrogen leakage, the gases will be diluted and ventilated outside, to maintain concentrations below combustible limits. Under most release cases, the ignition of hydrogen isotopes is not expected because of the elimination of ignition sources and the high ventilation rate.

ESTIMATED DOSE HAZARD		CODE CLASSIFICATION
MEMBER OF PUBLIC	PLANT PERSONNEL	
< 25 rem DUAL FAILURE LIMIT	< 25 rem DUAL FAILURE LIMIT	CLASS 1
< 500 mrem SERIOUS PROCESS FAILURE LIMIT		CLASS 2
	< 10 rem PLANNED SINGLE EXPOSURE LIMIT	CLASS 3
< 5 mrem 1% OF SERIOUS PROCESS FAILURE LIMIT	< 500 mrem ANNUAL LIMIT FOR A NON-ATOMIC RADIATION WORKER	CLASS 6

*1 rem = 10 m SIEVERTS

TABLE 1

GUIDE TO CODE CLASSIFICATION OF PROCESS SYSTEM BASED ON ESTIMATED DOSE HAZARD (INTERPRETED FROM CAN3-N285 O-M81)

The deuterium circuit, in the process system, has a number of automatic isolating valves, which will close to limit the escape or migration of tritium (Figure 2). Isolation will take place under a number of abnormal conditions, including power failure and detection of a gas release or fire.

DOSE AND RELEASE MODELING

In the remainder of this discussion, a number of schemes for modeling releases and estimating the doses that various groups of people could receive under accident conditions are reviewed.

Release and Rise of Hot Gases

When the plant is operating, the majority of the radioactive inventory is contained, as a liquid, in the distillation columns (Figure 2). When the plant is shut down and the system warmed up, the isolation valves will be shut and the majority of the inventory will be contained as a gas in the two expansion tanks. This places 200 000 Ci* as a gas at 300 kPa in the small expansion tank and 11 000 Ci as a gas at 450 kPa in the large expansion tank.

The release of tritium gas is a minimal radiological hazard until it is converted to an oxide. This conversion can take place long after the gas has been released from the building and is well diluted or it can take place more rapidly as a burn on release or as an

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* 1 Ci = 37 GBq

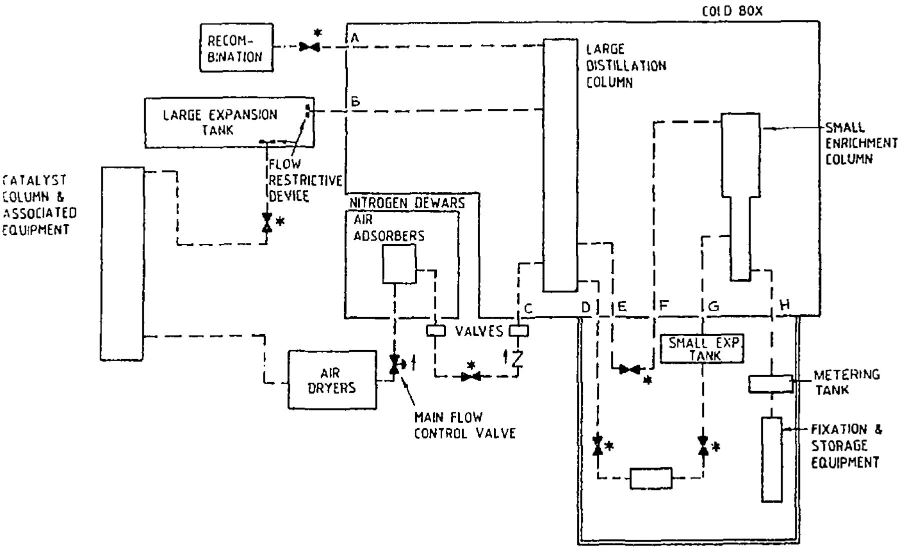

FIGURE 2

PROCESS SYSTEM SHOWING AUTOMATIC ISOLATING VALVES*, FLOW RESTRICTIVE DEVICES, MAIN CONTROL VALVE AND IMPORTANT CHECK VALVE.

explosion either inside the process system pressure boundary equipment or once released to the building. To consider a worse case hazard, it is assumed that any release of tritium is in the oxide form. These releases will be at elevated temperatures, due to the burn or explosion that takes place during the accident and will possess some buoyancy. The subsequent plume rise is dependent on the power of the burn and increases with quantity of hydrogen released for a given time. The higher the plume power, the greater the plume rise or theoretical release height, the wider the dispersion and the lower the maximum estimated dose.

One of the accidents considered is a release of the large expansion tank inventory through a pipe break at a point that cannot be isolated from the tank (Figure 2, points A,B,C). This would result in a release of 100 m^3 of hydrogen containing up to 11 000 Ci of tritium over an estimated 30 minute period based on "3/8 inch" orifice restrictive device in the tank outlet. From buoyant plume rise relations given in Appendix A of CSA-N288.2 [4], with a power from combustion of 1.44×10^5 cal/s of which an estimated 25 percent is lost due to radiant heat loss [2], buoyancy flux parameter (F) is 4.0 x m^4/s^3. These conditions give, for neutral conditions, a final plume rise above the 18 metre high building of

$$h_b = 1.6\, F^{1/3}\, U^{-1}\, (3.5\, x^*)^{2/2} \qquad (1)$$
$$= 12.1 \text{ m}$$

where $U = 5$ m/s and $x^* = 14\, F^{5/8}$

Under stable conditions, a final plume rise is

$$h_b = 2.4\, (F/U\, S)^{1/3} \qquad (2)$$
$$= 28.4 \text{ m}$$

where $U = 2$ m/s, $S = 1.2 \times 10^{-3}/s^2$ for Pasquill F weather.

Under stable conditions the theoretical release height would be approximately 46 m. The maximum concentration, under these conditions, occurs 3.5 km from the release point, where the land elevation is 8 m higher than the release point land elevation. This would give an estimated equivalent release height of 38 m and a resultant axial concentration [3] of 2.5×10^{-5} $(Ci/m^3)/(Ci/s)$.

Under neutral conditions the estimated release height of 30 m must be adjusted by the "modified 2½ times rule" in Bryant [3] for an 18 m building. The theoretical release height is then adjusted to 21 m. The maximum concentration under this condition occurs at a distance of 200 m from the release point. The land 200 m from the building houses other buildings of the laboratories and is approximately 10 m lower than the land elevation at the release point. With the final estimated release height of 30 m, the maximum concentration would be 3.2×10^{-5} $(Ci/m^3)/(Ci/s)$.

A release of 11 kCi with a concentration of 3×10^{-5} $(Ci/m^3)/(Ci/s)$ would result in an estimated dose equivalent of [4].

$$\text{Dose} = \frac{11.0 \text{ kCi} \times 3.0 \times 10^{-5} \text{ (Ci/m}^3\text{)/(Ci/s)}}{37 \text{ (Ci/m}^3\text{)/(rem/s)}}$$

$$= 9 \text{ mrem}$$

The maximum concentration of 7.0×10^{-6} at the plant boundary, 6 km away would occur under stable conditions. The estimated dose equivalent to a member of the public at the boundary fence would be 2 mrem. This would be approximately 20 times larger or 40 mrem if the 200.0 kCi were released along with the large hydrogen inventory.

Roof Vent Release

Greater, on site, doses would occur if the tritium inventory was released as an oxide, but slowly enough, such that the release height, above the building, is small and that the release is essentially roof top. When looking at concentrations of releases at locations close to the building, credit can be taken for the more rapid dilution that takes place in the turbulent mixing in the lee of the building [5]. The total dilution is the sum of the meteorological dilution at a specified distance away from the release source (Da) and the dilution due to turbulent mixing in the lee of the building (Db). For Pasquill F weather at 100 m, the diffusion coefficient is 1.7×10^{-2} (Ci/m^3)/(Ci/s). The equivalent dilution factor (Da), is $1/1.7 \times 10^{-2}$ or 59. The dilution factor Db is

$$Db = CAU \qquad (3)$$

where C is the size of the volume source in multiples of A (typically 0.5)
A is cross sectional area of building perpendicular to the wind (in the case of TEP, 1020 m^2)
U is the mean wind speed (2 m/s under Pasquill F weather conditions)

$$Db = 0.5 \times 1020 \times 2.0$$
$$= 1020$$

Total dilution is 1079 with a resultant diffusion coefficient of 1/1079 or 9.3×10^{-4} (Ci/m^3)/(Ci/s).

The estimate dose equivalent 100 m downwind of the building from a 200 kCi release is

$$\text{Dose} = \frac{200 \text{ kCi} \times 9.3 \times 10^{-4} \text{ (Ci/m}^3\text{)/(Ci/s)}}{37 \text{ (Ci/m}^3\text{)/(rem/s)}}$$

$$= 5.0 \text{ rem}$$

This dose diminishes to 2.8 rem and 1.4 rem at distances of 500 m and 1000 m downwind. Pasquill F weather conditions provided the maximum estimated doses at these locations.

Releases into Nitrogen Purged Glove Box

A glove box provides a secondary barrier around equipment that contains high concentrations of tritium gas. Accounting for equipment, the glove box volume is estimated to be 3.0 m^3. A nitrogen atmosphere, at slightly above atmospheric pressure, is maintained in the box by means of a nitrogen purge of 3.8 L/s from the boil-off of a liquid nitrogen tank and nitrogen cooled equipment. Any release into the glove box is assumed to be uniformly mixed with the nitrogen atmosphere and subsequently vented to the inner roof area, diluted with building ventilation and exhausted out the stack. The glove box atmosphere will be continuously monitored for any buildup of oxygen or inleakage of tritium. Detection of an abnormal condition will shut the isolation valves and sound an alarm.

The largest tritium release into the glove box takes place when a pipe break on a line to the small expansion tank occurs and the line cannot be isolated (Figure 2, points F,G and H). Flow restrictors in the form of "¼ inch" flow orifices may be used on the outlets of the expansion tank to reduce the exit flow rate during such a release. The release rate averages 16 L/s, with the expansion tank pressure reducing to essentially atmospheric pressure after 130 seconds. By modeling the glove box atmosphere, estimates of the concentration of hydrogen in the gas being released out the vent line can be made. The rate of change of the hydrogen in the glove box during the first 130 seconds can be described in terms of the grams of hydrogen entering and leaving the glove box as

$$\frac{dA}{dt} = \frac{16 \text{ L/s} \times 5 \text{ g/mole}}{22.4 \text{ L/mole}} - \frac{3.8 \text{ L/s} \times A \text{ g}}{3000 \text{ L}}$$

where A is the grams of DT in the box at any time t.

Integrating and determining the constant at $t = 0$, $A = 0$, the expression for the grams in the glove box during the first 130 seconds is

$$A(t) = 2819.55 - e^{7.94-0.00127t} \qquad (4)$$

At the time when the release into the glove box stops, t = 130 s, 439 grams of hydrogen (DT), gas is in the glove box. The hydrogen in nitrogen concentration is about 65 percent. This gas, when vented to the building, will go through a period when it will be combustible (Figure 3). The rate of change of hydrogen in the glove box, after the release from the small tank stops, is dependent only on the exit rate from the glove box due to the nitrogen purge and is described as

$$\frac{dA}{dt} = - \frac{3.8\ L/s \times A\ g}{3000\ L}$$

integrating and solving for the constant $t = 130$ s and $A = 439$ g, the expression for grams in the glove box with t 130 s is

$$A(t) = e^{6.25-0.00127t} \qquad (5)$$

PLOTTED FROM DATA PUBLISHED BY
H.F. Coward and G.W. Jones, Limits of Flammability of Gases and Vapours, Bulle 503, Bureau of Mines, U.S. 1952.

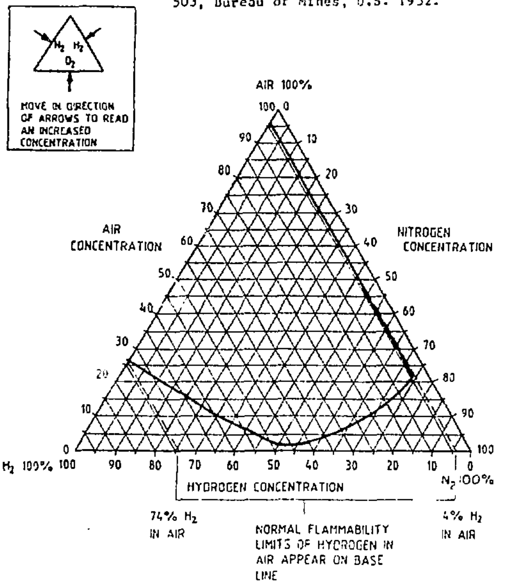

FIGURE 3
FLAMMABILITY LIMITS OF HYDROGEN-AIR-NITROGEN MIXTURES

The hydrogen in nitrogen will remain combustib when mixed with air as long as the concentrati remains above approximately 4%. This occu when A is greater than 26.8 grams and fr equation 5, occurs until t is 2338 s or about minutes. The time t the vent gas is combustib can be reduced to about 3.5 minutes if a 100 L purge was to be initiated at the 130 s point a would require about 20 m^3 of nitrogen gas.

This modeling assumes that the hydrogen gas mixed uniformly in the glove box and does n rise to the top of the box, where it would vented out sooner. This buoyancy effect wou lead to an increase in the exit rate of t hydrogen and reduce the concentration in t glove box at any time. With a buildi ventilation rate of approximately 4 m^3/s, t exit gas will be quickly diluted out of t combustible region before being vented of the stack.

Operator Hazards Inside Building

Operators, inside the building, can receive radiation doses in basically two ways. The first is through exposure to tritium oxide that has been released to the building air. The dose received is then sum of the doses due to inhalation and the absorption through the skin [4,6]. The second way is from the exposure due to skin wetting with tritiated heavy water that has leaked out of the process [6]. Heavy water is found in the first stage of the process and in the make-up and off-gas systems. The dose to the operator is dependent on the concentration levels but also on the time that the operator is subjected to the exposure situation. The time for an operator to exit the process building (Figure 4) from any point, on the main floor following a gas release, is estimated to be 21 seconds. This is composed of 10 seconds detection time, 6 seconds travel time (15 metres at 2.5 m/s) and 5 seconds to open and pass through the exit doorway to the link building.

A more probable location for a pipe failure and release to the process building is at a point where piping penetrates the cold box (points A,B and C, Figure 2). A pipe failure at one of these points, which cannot be isolated from the large expansion tank, would release the entire inventory of the large expansion tank. With the system warmed up, the initial pressure is approximately 5 atmospheres absolute. The initial release rate from the tank is 3.53 m^3/min. Added to this is the approximately 6 m^3 of gas that is stored in the equipment volumes and free to be released. This gives a release 9.53 m^3 of deuterium gas containing approximately 0.45 Ci/L (at 5 atmospheres) of tritium in the first minute after the failure. The release thereafter would be 3.53 m^3/min with continual reduction due to reduced tank pressure. The first minute tritium activity release would be 858 Ci. This would produce tritium in air concentration in the lower volume of the building, 1058 m^3, of 0.068 Ci/m^3 assuming the buoyancy of the hot release causes the release to move up the tower, above the main floor, after 5 seconds. The average first minute concentration would be equivalent to 3.1×10^3 derived air concentrations (DAC) (1 DAC = 22 Ci/m^3) [7] and for the 21-second exposure give a dose of:

$$\text{Dose} = \frac{2.5\ \text{mrem}}{\text{DAC}\cdot\text{h}} \times 3.1 \times 10^3\ \text{DAC} \times \frac{21\ \text{s}}{3600\ \text{s/h}}$$

$$= 45\ \text{mrem}$$

A release of the tritium inventory of the small expansion tank to the lower section of the building is not expected for a number of reasons. The tank is enclosed in the glove box with small leaks being vented out to the top of the building and not exposing operators in the

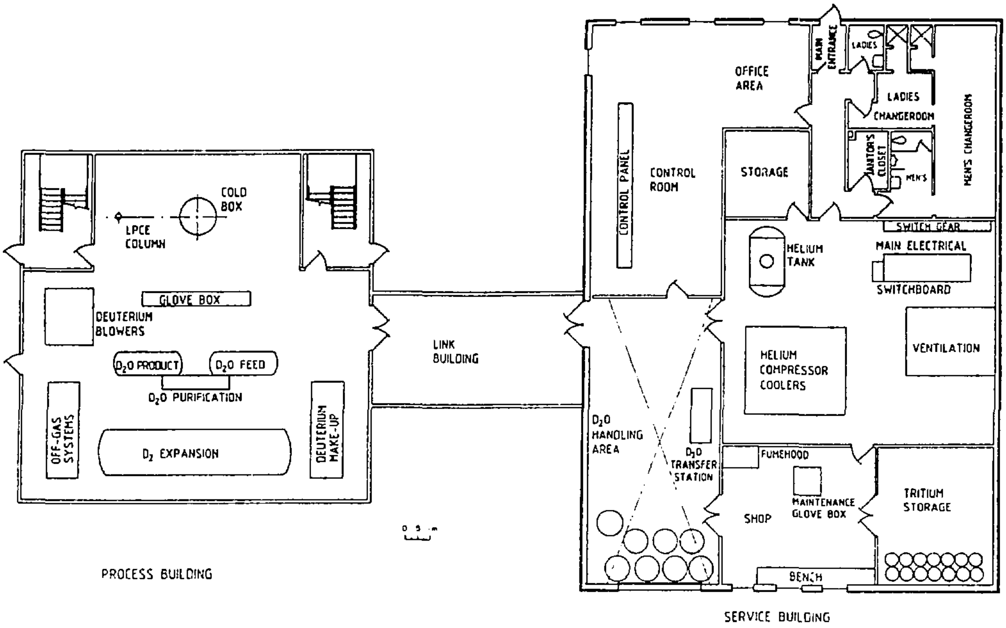

FIGURE 4 TRITIUM EXTRACTION PLANT FLOOR PLAN

building. The glove box air is monitored for oxygen and tritium and would warn operators of any abnormality. The tank design pressure is 2.0 MPa and should withstand an internal explosion initiated at atmospheric pressure. Oxygen migration into the system is monitored, reducing oxygen accumulation and the chances of an internal explosion. The nitrogen atmosphere in the glove box would exclude inleakage of air (oxygen) into the equipment when their internal pressure is below atmospheric pressure. The tank is located on the mezzanine level, an infrequently occupied area. Serious exposure to operators on the main floor is very unlikely, as the combusted gas would be above the main floor and have a tendency to move up rather than down.

A failure of the large expansion tank and instantaneous release of the 11 kCi of tritium would produce a high tritium in air concentration but at the same time the hot gases would likely blow out the building blowout walls and would rise quickly up the tower with fresh air entering at the bottom. Dose estimates for such a case are approximately 500 mrem, but physical injury would also be likely.

Streaming of small releases are more difficult to model. Uniform mixing has been adopted in the dose calculation here. An operator leaving the area would likely move into or out of the stream and the average balance out.

Dose From D_2O Skin Wetting

The committed dose due to wetting of skin can be large yet controlled in three ways. The first is to reduce the area of contact with the skin by protecting the skin with a barrier. A very practical barrier to be used in the TEP is the rubber glove. A second reduction can be accomplished by the removal of the cotton coveralls that have absorbed HTO - contaminated vapour. This action can reduce exposures of shorter than 10 minutes to about half of what would be expected had the coveralls not been removed[6]. The third step to reduce the dose due to skin wetting is to wash the skin area as quickly as possible after being wetted. Washing within a minute of a two minute exposure can reduce the net intake of tritium by more than half[6]. (Other techniques, such as increased fluid intake, can help to reduce the committed dose after the fact, but will not be considered here.)

If a plant operator has part of his body wetted with tritiated heavy water, it is important that he remove the contaminated clothing and shower as quickly as possible. The showers in the TEP are located in the service building (Figure 4). The longest travel distance is from the process building. By considering estimated times (i.e. walking, opening and passing through doorway, showering, etc.), for the specific operation it is possible to predict the time for an operator, wetted with tritiated heavy water, to take a shower.

The approximate time to travel the 50 metres, through 5 doors and take a shower after being wetted on the main floor (the most likely location) is 1 minute. The time for an operator to travel 15 metres and exit the door into the link building and reach fresh air is 21 seconds. Included in these times is 10 seconds for the operator detection of the operator detection of the event.

The dose to an operator can be estimated by making some assumption regarding the area of the skin that is wetted. According to Reference Man [8], the area of the head, shoulder and torso is about 45% of the estimate total skin area of 18 000 cm^2. Assuming that ¾ of this area gets wetted, from Bush [6], a 6 000 cm^2 wetting for 1 minute would result in an intake of approximately 2.5 mCi and give a committed dose of

$$\text{Dose} = \frac{2.5\ \text{mCi}}{43\ \text{L}} \times \frac{1000\ \text{Ci}}{\text{mCi}} \times \frac{4\ \text{mrem}}{\mu\text{Ci/L}}$$

$$= 233\ \text{mrem}$$

The dose from a heavy water wetting incident will also include a committed dose due to the tritium in air contamination. The intake is a combination of both adsorption through the skin and inhalation. From experience at CRNL, the measured tritium in air contamination, in adequately ventilated locations, as is the TEP building, never exceeds 250 DAC. Assuming that the level is at a maximum of 500 DAC, then the committed dose for the 21-second exposure - the time to reach the link door and fresh air, would be

$$\text{Dose} = \frac{2.5\ \text{mrem}}{\text{DAC-h}} \times \frac{21\ \text{s}}{3600\ \text{s/h}} \times 500\ \text{DAC}$$

$$= 7.3\ \text{mrem}$$

In documented incidents at CRNL, parts of the head, shoulders and arms of an operator were wetted while on another operator parts of the head, shoulders and torso were wetted during a heavy water spill incident. For comparison purposes, the actual dose due to this incident for the two operators in question was estimated to be 250 mrem, based on the increase in tritium level in urine samples. The heavy water involved in these spills was reactor grade, with about 10 Ci/L tritium contamination. The TEP estimated dose has good agreement with these actual incidents. The initial feed into the process will likely be greater than 20 Ci/L but should be reduced to 10 Ci/L or less when the bulk of NRU water is processed. (This is expected to take less than two years). If the tritium concentration in this accident had been 20 Ci/L, rather than 10 Ci/L, the dose to the operator could have been about 500 millirem.

SUMMARY

Approval of the code classification by the AECB has been obtained. All equipment in the process has been classed as class 6 with the exception of the small expansion tank located in the glove box which has been classed as class 2. Some extra requirements have been established for the large expansion tank and critical piping entering the cold box. Design and construction of the plant is proceeding with a tentative start up date of late 1986.

The models and techniques outlined in this paper will be expanded and combined with probability analysis to determine the overall plant risk. This will form the major part of the next step in licensing, the FSAR. Operating and commissioning procedures will also be included in the FSAR.

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