A COMPACT, LOW COST, TRITIUM REMOVAL PLANT FOR CANDU-6 REACTORS

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1.0 INTRODUCTION

Tritium concentrations in CANDU-6 reactors are currently around 40 Ci/kg in moderator systems and around 1.5 Ci/kg in primary heat transport (PHT) systems. It is expected that tritium concentrations in moderator systems will continue to rise and will reach about 80 Ci/kg at maturity. A more detailed description of the increase in tritium concentrations in the moderator and PHT systems of CANDU-6 reactors is given in the next section of this paper. While moderator systems currently contribute more than 50% to tritium emissions, the impact of acute releases of moderator water is more severe at higher tritium concentrations. This impact can be substantially reduced by the addition of an isotope separation system for lowering the tritium level in the moderator system. In addition, lower tritium levels in CANDU systems will inevitably result in reduced occupational exposures, or will provide economic benefits due to ease of maintenance because less protective measures are required and maintenance activities can be more efficient.

New technology has been developed at Ontario Hydro over the past 15 years, which can greatly reduce the cost and size of a Tritium Removal Plant (TRP) as compared to Ontario Hydro's Darlington Tritium Removal Facility (DTRF) which is designed to reduce the tritium level in the moderators of Ontario Hydro's 20 CANDU reactors to near 10 Ci/kg. In addition, excellent engineering, schedule, and cost data have been developed as a result of the successful completion of the design, construction, installation and commissioning of a Tritium Purification System (TPS) for the Princeton Plasma Physics Laboratory.[1,2,3] Ontario Hydro has design responsibility for the isotope separation system for the International Thermonuclear Experimental Reactor (ITER)[4] project. The isotope separation system will separate and concentrate the tritium in this proposed multi-billion dollar international fusion reactor. ITER design responsibilities and work on the Princeton TPS have spurred millions of dollars of R&D and design and led to significant advances in cost and tritium inventory reduction.

The compact TRP's described in this paper have been sized to serve a single CANDU-6 unit, but the systems described can also be enlarged or deployed in a modular fashion to serve a multi-unit station like Wolsung. The TRP design consists of a low-inventory, leak-tight electrolysis cell or group of cells in the front-end to convert the D_2O (mixed with HDO and DTO) from the moderator system to D_2, HD, or DT. This is followed by a cryogenic distillation back-end system which is sufficient to enrich the moderator tritium to between 50% and 80% DT while detritiating the bulk of the D_2 gas which is then converted back to heavy water by recombining the D_2 with oxygen. This compact Cryogenic Distillation (CD) System is much simpler than the 4-column Princeton TPS which processes hydrogen isotopes from the spent fuel of the Princeton fusion reactor and produces pure T_2 with a total tritium inventory in the isotope separation system of only about 1 gram. It is also simpler and very much smaller than the 4-column Darlington TRF which lowers the tritium inventory in Ontario Hydro's CANDU reactors. For example, the Darlington-TRF CD contains 2300 m^3 of hydrogen, deuterium, or tritium while the CD for the CANDU-6 TRP contains only about 8.5 m^3 of hydrogen isotopes. Allowing for the fact that Darlington TRF services not one, but 20 reactors, the inventory in the CANDU-6 TRP is still 13.5 times smaller per reactor.

This compact CANDU-6 TRP also offers the advantage of on-site detritiation, and hence avoids the cost, infrastructure, and labour necessary for shipping of heavy water to an off-site processing facility. It is small enough to fit into an existing building and it can reduce C-14 production and C-14 release by up to 30% to 40% by using natural oxygen in the TRP recombiner to displace the enriched O-17 in the moderator heavy water which is responsible for C-14 production. The TRP can also provide on-line upgrading of the moderator water at a very low incremental cost by stripping H from the D_2 gas and raising the isotopic analysis of moderator heavy water from 99.9%D to 99.95%D. Such an increase can reduce fuel burn-up by about $25K/year per reactor, as well as offering additional flexibility, particularly for stations equipped only with a single upgrader.

Since the proposed CANDU-6 TRP design is simple, this means that process control implementation is easier, and the fewer components will result in lower maintenance requirements and lower operating costs. The compact CANDU-6 TRP can be confidently designed and built over a two year schedule, subject to licensing and station interface issues. This is a result of experience with the more complicated Princeton TPS system. Ontario Hydro has developed unique design tools such as the FLOSHEET[5] and DYNSIM[6] simulation codes, which enable fast and accurate design optimization for any tritium separation design task.

2.0 TRITIUM CONCENTRATIONS AND ISOTOPICS IN CANDU-6 REACTORS

Figure 1 shows tritium activity growth in the moderator systems of Embalse, Pt. Lepreau, Wolsung-1 and Gentilly-2 reactors. Tritium concentrations have increased steadily since reactor start-up, and in January 1996 were at an average of about 40 Ci/kg in moderator systems.[7]

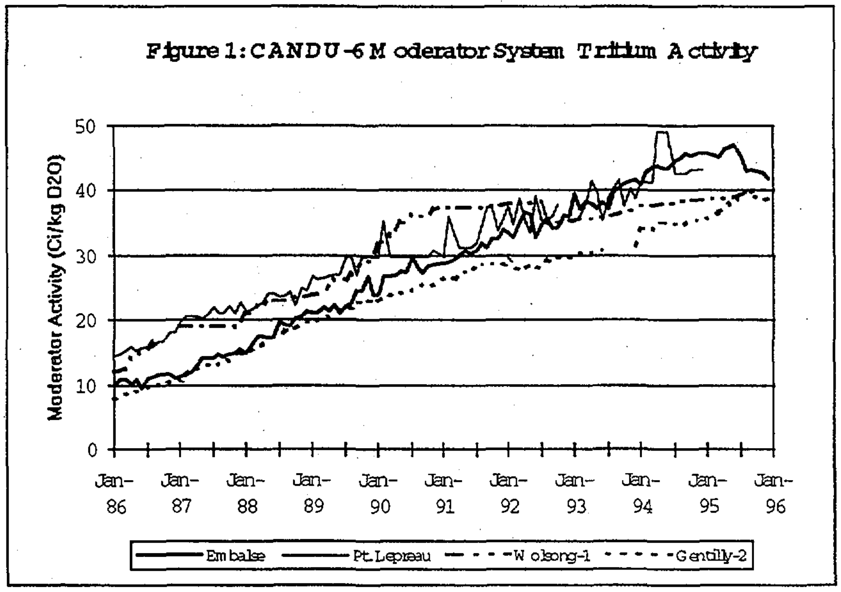
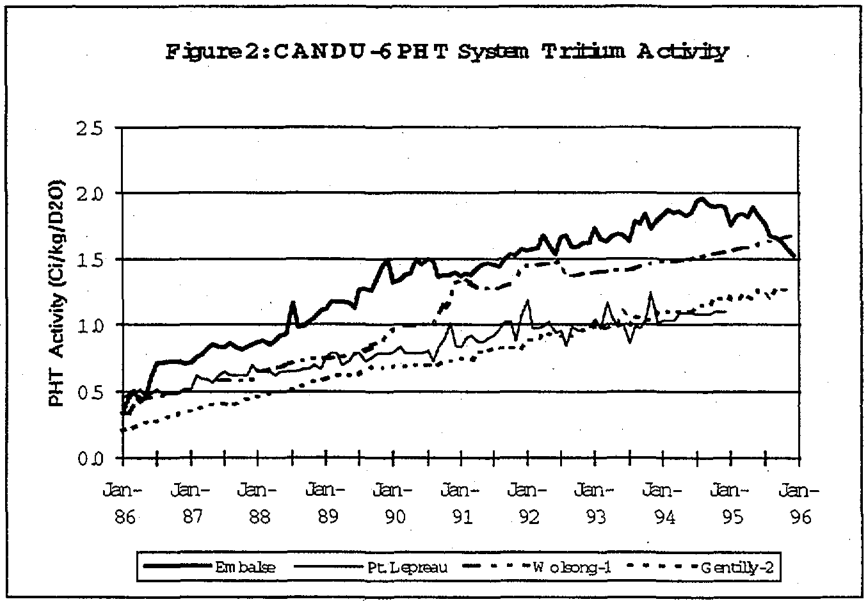
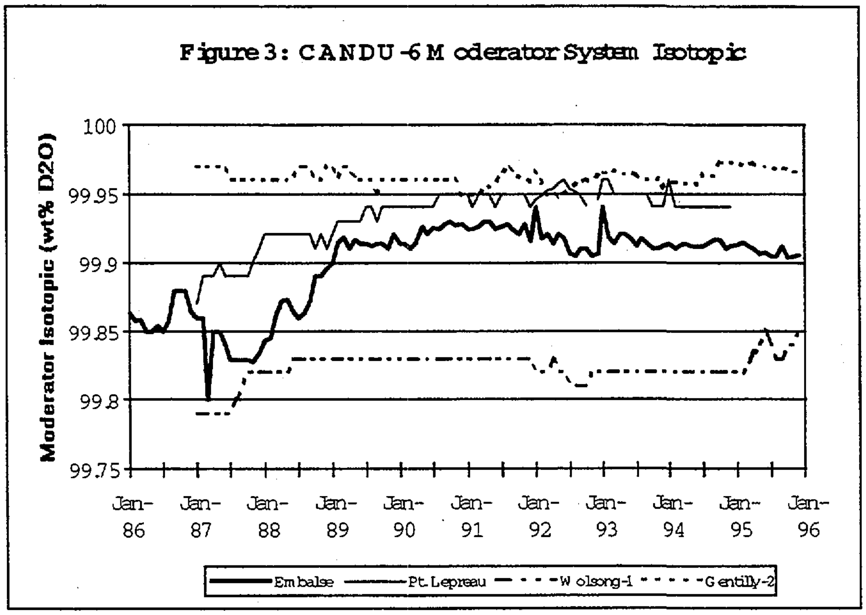


Figure 2 shows the tritium activity in the Primary Heat Transport (PHT) systems of the same reactors.[7]



In January 1996, PHT concentrations averaged around 1.5 Ci/kg. It is expected that tritium concentrations in moderator systems will continue to rise and will reach about 80 Ci/kg at maturity.

In contrast with tritium concentration, moderator heavy water concentrations display a relatively large scatter and are shown in Figure 3.[7]



While moderator systems are estimated to contribute somewhat more than 50% to tritium emissions, the impact of an acute release becomes more severe the higher the tritium concentration. The data presented in Table 1 suggest that tritium emissions which are significant fractions of total emissions at CANDU-6 reactors could be significantly reduced, if detritiation of moderator systems is performed.

Table 1[8]
Moderator Contribution to Tritium Emissions

Year	**Contribution to Total Emissions**			**Contribution to Airborne Emissions**		
	Gentilly 2	Point Lepreau	Wolsung 1	Gentilly 2	Point Lepreau	Wolsung 1
1988	43%	70%	71%	42%	79%	68%
1989	25%	81%	70%	9%	77%	67%
1990	70%	68%	54%	68%	55%	49%
1991	85%	65%	58%	86%	59%	51%
1992	69%	78%	59%	59%	92%	58%
Average	58%	73%	62%	53%	72%	59%

3.0 TECHNOLOGY DEVELOPMENTS SINCE DARLINGTON TRF DESIGN AND CONSTRUCTION

New technology has been developed at Ontario Hydro over the past 15 years, which can greatly reduce the cost and size of a Tritium Removal Plant (TRP) as compared to the Darlington Tritium Removal Facility (DTRF).

In the past few years, Ontario Hydro has been the lead designer for the Isotope Separation System (ISS) for the International Thermonuclear Experimental Reactor (ITER)[4]. The ITER-ISS is a large 4-column cryogenic distillation system, with a stringent limit on tritium inventory. Efforts to reduce inventory, cost and size for the ITER-ISS have been successful, using an integrated design and R&D approach. Ontario Hydro has developed steady state and dynamic simulation codes (FLOSHEET and DYNSIM) for isotope separation systems. These have been used to analyze and optimize design configurations. An extensive R&D program has also been carried out at Ontario Hydro Technologies to test and characterize various column packings, hardware components, and process control technology for cryogenic distillation. Testing, development, and design work on the ITER fusion reactor's isotope separation system has reduced the column height by approximately 250% and the tritium and hydrogen isotope inventories by about 500% over the last three years.

This capability has been further strengthened due to the recent successful design, construction, testing, delivery, commissioning, and operation of the Tritium Purification System (TPS) for the Tokamak Fusion Test Reactor (TFTR) located at Princeton, New Jersey. The TPS is a 4-column distillation system integrated with a front-end palladium diffuser purifier. The TPS is designed to separate 99% pure tritium from all plasma exhaust gases. The TPS project was executed over a tight 17 month schedule, with a stringent operating inventory constraint of 10,000 Ci in the system. The TPS has operated reliably and consistently produced pure tritium product from the plasma exhaust gases. This tritium has been recycled for use in new fusion experiments instead of having to be disposed of as a nuclear waste. Operation of the Princeton's TPS has also provided a demonstration of improved computer control of CD systems.[9] Work on ITER and the TPS has spurred millions of dollars of R&D and design effort in cost and tritium inventory reduction. In addition, excellent engineering, schedule and cost data have been developed for the production of new, efficient, cryogenic distillation systems.[1,2,3]

4.0 PROCESS OPTIONS FOR THE COMPACT CANDU-6 TRP

The CANDU-6 TRP design consists of a front-end process for phase transfer of tritium in the heavy water to a D_2/DT stream followed by a CD system. A number of options are available for the front-end process:

OPTION	COMMENTS
Vapour Phase Catalytic Exchange (VPCE)	Used at Darlington, requires a number of discrete stages each involving deuterium gas and steam separation.
Liquid Phase Catalytic Exchange (LPCE)	Continuous column, hence more compact than the VPCE, but requires bigger CD than with other options.
Electrolysis (DE)	Requires smaller CD than for VPCE or LPCE front-ends. C-14 reduction possible.
Combined Electrolysis and Catalytic Exchange (CECE)	Smallest CD size. Combination of DE plus LPCE. Needs more electrolysis cells than DE. Electrolysis cells need to operate at higher tritium concentration than DE.

All of the above front-end options are technically feasible. However, the following general trends apply:

Floor area required: LPCE < DE & CECE < VPCE

Building height required: DE < LPCE < VPCE & CECE

Size of CD system: CECE < DE < VPCE < LPCE

C-14 Reduction Possible: DE & CECE

On this basis, electrolysis has been selected as a good compromise due to low space requirement, reasonable size of the associated CD system and the additional benefit of C-14 mitigation. This is not meant to exclude other options from consideration. Use of the compact CD will reduce the cost of any tritium separation system irrespective of the front-end option selected.

5.0 DESIGN OF THE COMPACT CANDU-6 TRP

The compact TRP has been sized to serve a single CANDU-6 unit, but can also be enlarged or deployed in a modular fashion to serve a multi-unit station like Wolsung. A representative flowsheet for the compact CANDU-6 TRP is shown in Figure 4.

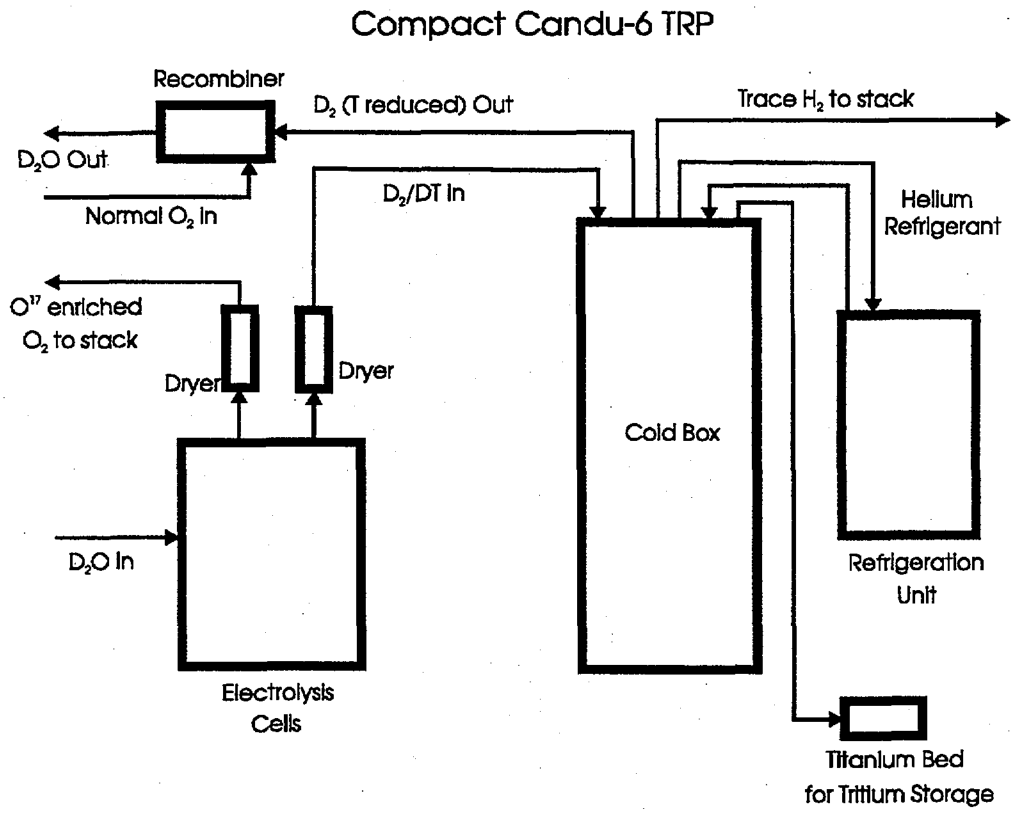


Figure 4: Compact CANDU-6 TRP Flowsheet

The TRP consists of an electrolysis front-end, utilizing low-inventory, leak-tight electrolysis cells of the type being tested at Atomic Energy of Canada Ltd. (AECL), Chalk River. The oxygen stream is dried to prevent water and tritium escaping to the environment and then discharged to stack. The D_2/DT stream is purified via an ambient temperature drier followed by an adsorber at 77K temperature (or a Palladium diffuser) and sent on to the cryogenic distillation (CD) system. This prevents water and any non-hydrogen gases such as oxygen from reaching the CD system. The CD system is designed to enrich tritium to between 50% DT and 80% DT as a low volume product. This can then be immobilized on titanium for storage. While CD units can readily be designed to produce pure tritium, the main design requirement for CANDU-6 reactor detritiation is to lower the tritium in the moderator heavy water. Enrichment to 80% DT (instead of 99% T_2) is sufficient to reduce the tritium containing stream to a very small volume without requiring the addition of additional column length, additional processing hardware, and a large amount of extra tritium inventory. A compact CANDU-6 TRP capable of holding the moderator water tritium concentration to below 10 Ci/kg, requires only 8 titanium tritide containers per year to hold the removed tritium product. Each container is about 17 cm diameter by about 1 m high.

The Darlington TRF uses a liquid hydrogen refrigeration system to provide cooling for the CD system, while the CANDU-6 TRP uses a low temperature gaseous helium refrigerator instead. This CANDU-6 CD system is much simpler to operate than the 4-column Princeton TPS, which, nevertheless, has demonstrated good operating performance in processing fusion reactor waste.

Figure 5 shows a plan view of the CANDU-6 TRP capable of holding CANDU-6 moderator water at 10 Ci/kg. It fits easily into a space which is 10 m by 8 m. The space is less for a TRP designed to hold CANDU-6 moderator water at 20 or 30 or 40 Ci/kg. These new small footprints and the much lower deuterium and tritium levels in the CANDU-6 TRP when compared with conventional CD technology mean that it should be possible to insert a CANDU-6 TRP into the reactor buildings rather than in a completely separate building such as used by Ontario Hydro's Darlington TRF.

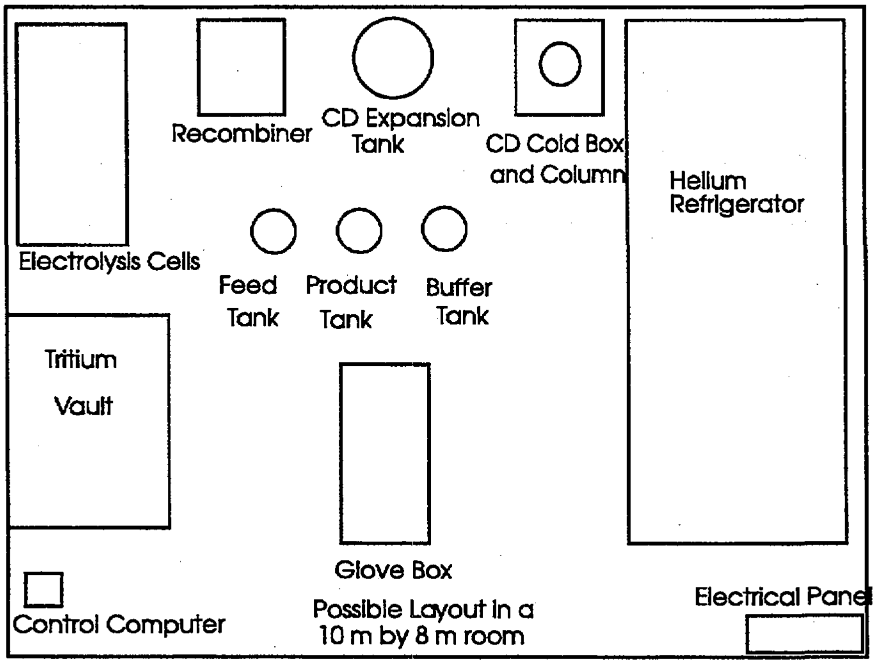


Figure 5: CANDU-6 TRP Layout

Figure 6 shows the inverse relationship between the target equilibrium concentration of tritium in the moderator and the required feed flow rate to the CANDU-6 TRP.

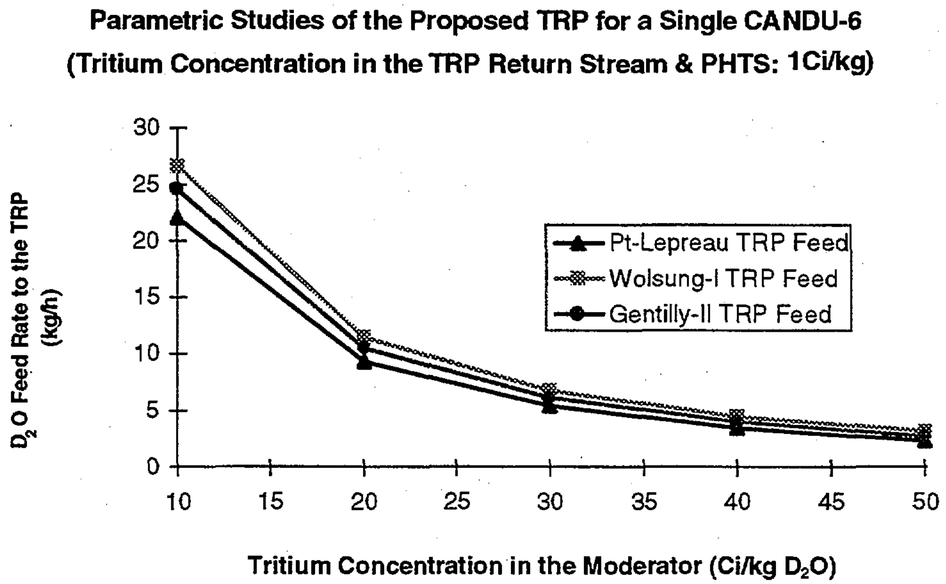


Figure 6: **Process flow vs. Moderator Ci/kg**

The elevation view of the compact CANDU-6 TRP is also very much smaller than the elevation view of the Darlington TRF (Figure 7). Darlington has two cold boxes, a large one over 30 m tall and 1.3 m in diameter and a smaller one 11 m tall and 1.23 m in diameter (part of this cold box is reduced to 0.6 m diameter). The cold box is a vacuum vessel which contains the isotope separation columns which operate just above 20 K and which must be completely insulated from the 300 K environment of the nuclear power station. The compact CANDU-6 TRP has a cold box which is 5.5 m tall (or shorter if holding the moderator water to 10 Ci/kg is not required) and 0.5 m in diameter. The magnitude of the size difference is really understood when the hydrogen isotope inventory in the Darlington system (2300 m^3) is compared to the hydrogen isotope inventory of the compact CANDU-6 TRP (8.5 m^3). The reduced size is an indication of a large cost reduction, while the reduced hydrogen inventory significantly reduces the consequence of any postulated accidental event.

The compact CANDU-6 TRP offers the advantage of on-site detritiation, and hence avoids the cost (including heavy water inventory cost to replace the inventory that is off-site), infrastructure, and labour necessary for shipping of heavy water to an off-site facility.

Implementation of a CANDU-6 TRP can allow a reactor the added benefit of reducing the production and hence the release of C-14 from the reactor. In addition, the TRP can be used for on-line upgrading of the moderator heavy water. Both of these benefits are available at only a small incremental cost.

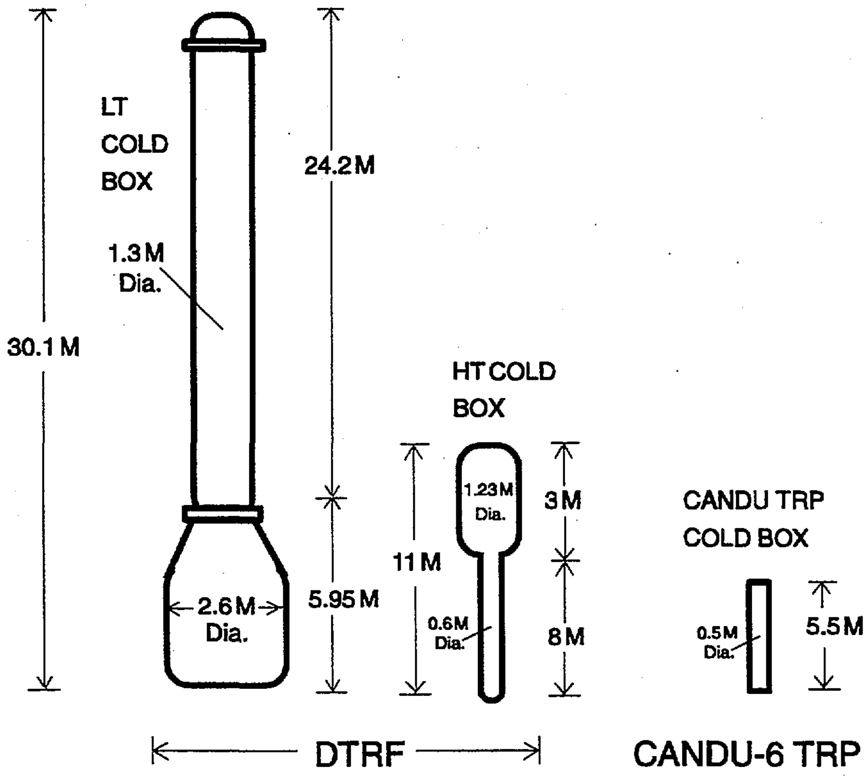


Figure 7: Elevation view

The CANDU-6 TRP can reduce C-14 production and C-14 release by up to 30% to 40%. It does this by using natural oxygen in the TRP recombiner which effectively displaces the enriched O-17 in the moderator heavy water. Oxygen-17 is enriched in the process of converting lake water to heavy water. It gets further enriched in the D_2O upgrading process. Whereas the natural abundance of O-17 is about 300 ppm in light water, the typical concentration in a moderator is about 550 ppm. The major source of carbon-14 in CANDU reactors comes from neutrons bombarding the oxygen-17 nuclei in the reactor moderator. With the absorption of a neutron and the ejection of an alpha particle, carbon-14 atoms are formed. Operation of the CANDU-6 TRP will gradually reduce the amount of O-17 in the moderator and thus gradually reduce the rate of carbon-14 production. Production rate changes due to the operations of the two sample compact CANDU-6 TRP designs (described below) is shown in Figure 8. Thus, it is possible to reduce C-14 emissions from a reactor by about 30% to 40% over 5 years at a low additional cost.

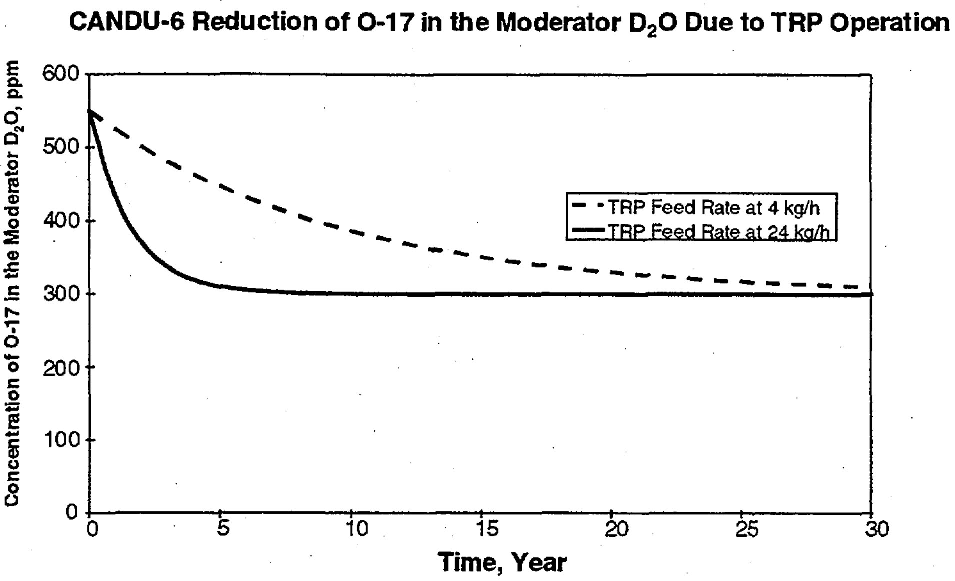


Figure 8: Reduction of O-17

Compact TRP technology can also be modified to provide on-line deuterium upgrading of the moderator water at a very low incremental cost. For example, the deuterium concentration in the moderator can be gradually raised from 99.9% to 99.95% by modifying our standard compact CANDU-6 TRP design to lower the concentration of H in the heavy water. Such an increase in the D level in the moderator can reduce fuel burnup by about $25K/year per reactor, as well as offering additional flexibility, particularly for stations such as Pt. Lepreau, equipped only with a single upgrader. Even for stations with two upgraders, the additional flexibility of on-line upgrading in the TRP can free up upgrading capacity to treat backlogs of D_2O stored in drums, or can allow the upgraders to work to higher product specifications.

Since the proposed CANDU-6 TRP design is simple, this means that process control implementation is easier, and the fewer components will result in lower maintenance requirements and lower operating costs. The CANDU-6 TRP can be confidently designed and built over a two year schedule, subject to licensing and station interface issues. This is a result of experience with the more complicated Princeton TPS. Ontario Hydro has unique design tools such as the FLOSHEET[5] and DYNSIM[6] simulation codes, which enable fast and accurate design optimization.

Although our design tools allow us to design any size cryogenic distillation system for any design requirement, two different sizes for the compact CANDU-6 TRP will be presented - a small system to hold the moderator at a concentration of 40 Ci/kg and prevent it from rising any further, and a larger TRP to bring the moderator concentration down gradually to 10 Ci/kg and hold it at this value. The parameters associated with the two sizes are shown in Table 2.

Table 2
TRP Parameters

Moderator Concentration at Maturity	40 Ci/kg	10 Ci/kg
TRP D_2O Feed Flow	4 kg/h	24 kg/h
E-Cell Power	30 kW	180 kW
E-Cell Tritium Inventory	0.3 g	0.4 g
CD Column Condenser Load	420 W	2100 W
CD Column Height	3 m	4.5 m
CD Tritium Inventory	2.7 g	4.7 g

It is envisaged that the larger CANDU-6 TRP will fit comfortably into a floor space of 10m by 8m. A smaller TRP will require less space. Commercial helium gas refrigeration systems are available in the desired sizes, and will also fit into the space required.

6.0 SAFETY AND LICENSING CONSIDERATIONS

Table 3 shows that the CANDU-6 TRP is estimated to have greatly reduced hydrogen and tritium inventories when compared to the Darlington TRF. Thus, the Darlington TRF can be viewed as an upper bound and a precedent for licensing of such a facility. Analyses similar to those used for licensing the DTRF can be used for the CANDU-6 TRP, and it should be easy to demonstrate that the facility can be engineered to be safe, both from the conventional and radiological safety viewpoint.

For example, in a room size of 10m x 8m x 6m high, it is not possible to reach the lower flammability limit of hydrogen in air (4 vol. %), even if the full 8.5 m^3 hydrogen inventory of the larger CANDU-6 TRP is released as the result of a postulated accidental event.

Table 3
Comparison of TRP Sizes, Inventories

	40 Ci/kg TRP	10 Ci/kg TRP	DTRF
D_2O Feed Flows kg/h	4	24	360
Refrigeration Watts	420	2,100	100,000
Column Height	3 m	4.5 m	30 m
H_2,D_2 Inventory	2 m^3	8.5 m^3	2300 m^3
Tritium Inventory	2.7 g	4.7 g	35 g

7.0 CONCLUSIONS

New compact cryogenic distillation technology can greatly reduce the size and cost of tritium decontamination equipment for CANDU-6 reactors. Compact new systems containing only 0.08% to 0.35% of the hydrogen inventory of conventional technology make it possible for every CANDU-6 reactor utility to consider using detritiation technology. With reduced tritium levels in the moderator heavy water, fewer maintenance operations will require plastic suits and plant tritium emissions will be reduced.

With fairly minor modifications, compact cryogenic distillation technology with a electrolysis front end can be used to supplement station moderator upgrader capability and can reduce carbon-14 production by about 30 to 40%. Although the process discussion deals mainly with an electrolysis front end, if it is necessary to use VPCE (vapour phase catalytic exchange), LPCE (liquid phase catalytic exchange), or CECE (combined electrolysis and catalytic exchange), all of these front ends require cryogenic distillation as a back end. The cost of all of these approaches to tritium isotope separation will be significantly reduced if compact cryogenic distillation is used instead of conventional distillation technology.

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[2] A. Busigin, C.J. Busigin, F. Adamek, K.B. Woodall, J.R. Robins, D.G. Bellamy, C. Fong, K.M. Kalyanam, S.K. Sood. "Control system implementation for a complex low inventory cryogenic distillation system for Princeton TFTR." SOFT 18 Fusion Conference Proceedings, Fusion Technology, Vol. 2, pg 1051, (1994).

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[5] A. Busigin and S.K. Sood, "Process Simulation with Microcomputers: A Generalized Simulation Code for Application to Fission and Fusion Systems", Bulletin of the Canadian Nuclear Society, Vol 9, No. 3, May, June 1988.

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[7] F. Fusca, K. Kalyanam, "CANDU Owners Group - CANDU Station Chemistry Newsletter 97-01, D_2O Management for 1995", April 1997.

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CNS Technical Program

WEDNESDAY, 1997 June 11

Session 4C **Applied Nuclear Research and Development** **Leaside Room**

Chair: K. Weaver
Ontario Hydro

09:00-09:25	*Qualification of Passive Autocatalytic Recombiners for Post-LOCA Hydrogen Mitigation in CANDU® Stations* by J.V. Loesel Sitar, R. Moffett, G.W. Koroll, W.A. DeWit (Atomic Energy of Canada Limited)
09:25-09:50	*Determining the Effect of Turbulent Shear on Containment Aerosol Dynamics Using Microgravity Experiments* by C.K. Scott, M. Abdelbaky (Atlantic Nuclear Services Ltd.)
09:50-10:15	*The Carbon Filter Programme: An Example of Success in Applied Nuclear R&D* by J.C. Wren (Atomic Energy of Canada Limited) and K.R. Weaver (Ontario Hydro)
10:15-10:45	Break
10:45-11:10	*Effects of Gamma and Thermal Neutron Radiation on Nitrocellulose* by K. Heppell-Masys, H.W. Bonin, V.T. Bui, M. Bickerton, D. Murphy (Royal Military College of Canada)
11:10-11:35	*Fission Products Measuremetns in the Slowpoke-2 Reactor at the University of Toronto* by S. Sonoc, G.J. Evans (University of Toronto)
11:35-12:00	*Engineering Progress of CNS Concept in HANARO* by C.O. Choi, K.N. Park, S.H. Park, J.M. Sohn, M.S. Cho (Korea Atomic Energy Research Institut)