



Conference Paper

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DETRITIATION PROJECT AT
CHALK RIVER
LABORATORIES**

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A LIGHT-WATER DETRITIATION PROJECT AT CHALK RIVER LABORATORIES

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The NRU reactor rod bays is a large, open pool of water that receives hundreds of fuel rods annually, each carrying a small amount of residual tritiated heavy water. The tritium concentration of the rod bays water has risen over the years, to a level that is of concern to the operations staff and to the environment. The proposed long-term solution is to reduce the rod bays tritium concentration by direct detritiation of the water.

The Combined Electrolytic-Catalytic Exchange (CECE) process is well suited to the light-water detritiation problem. With a tritium-protium separation factor greater than five, a CECE detritiation process can easily achieve the eight orders of magnitude separation required to split a tritiated light-water feed into an essentially tritium-free effluent stream and a tritiated heavy water product suitable for recycling through a heavy water upgrader.

This paper describes a CECE light-water detritiation process specifically designed to reduce the tritium concentration in the NRU rod bays to an acceptable level. The conceptual design of a 600 Mg/a detritiation process has been developed and is now at the stage of project review and the beginning of detailed design.

I. INTRODUCTION

Heavy water reactors produce tritium in the moderator and coolant by deuterium neutron capture and this builds up with time. Transfer of fuel out of the reactor core provides a critical part of a path for tritium to be transferred to the work areas and the environment. At AECL's National Research Universal (NRU) reactor at Chalk River the transfer of spent fuel rods to the rod bays has resulted in a significant buildup of tritium in the rod bays water. Largely because of the procedures used during medical isotope production at NRU, the modest concentration of tritium in the open pool of the rod bays constitutes a significant source of worker dose. The rod bays are also old and not leak-tight, and thus provide the final link in a path for tritium to leak to the environment. AECL is applying efforts to reduce the tritium transfer rate at all points along the release route. Continuous removal of tritium from the reactor core coolant and from the rod bays appear to be the most practical means of

reducing the tritium in the rod bays. This paper describes AECL's proposed system for removing tritium directly from the rod bays water.

II. NRU DETRITIATION STRATEGY

The NRU rod bays contain about 1070 Mg of pure water and the tritium concentration is currently about 10 mCi/L. Tritium is introduced to the rod bays in residual reactor heavy water on each rod transferred from the reactor. Tritium is lost from the rod bays by decay, evaporation and water leakage to the ground. The concentration in the bays will depend on the balance of all these variables. A mathematical model has been set up to predict the time-profile of tritium given the operating and environmental conditions. Given a target tritium concentration, the appropriate detritiation rate can be determined with a simple extension to this model.

There is a practical limit to how far detritiation can lower the rod bays tritium concentration. Like all purification problems where there is a constant source term (e.g., constant flow of tritium to rod bays), there are diminishing returns with the increasing size of the purification plant. Every halving of the rod bays tritium concentration by detritiation requires a near doubling of the detritiation plant size. Since the objective is to minimize worker dose (from inhaled vapor) and tritium entering the groundwater at a reasonable cost, it was decided to target a long-term rod bays tritium concentration of not more than 1.5 mCi/L. This would result in a full step reduction in the tritium hazard category (achieving about 1 $\mu\text{Ci}/\text{m}^3$ in air) and be about 1/10th the rod bays steady-state tritium concentration with no detritiation. This is approximately the same degree of detritiation as is achieved in the moderator of most CANDU reactors.

Detritiation of the NRU moderator or the rod bays water will both achieve reduced tritium concentrations in the rod bays. However, it has been determined to be impractical to detritiate either the moderator or the rod bays alone to the level required to achieve the target of 1.5 mCi/L of tritium in the rod bays. It follows, therefore, that detritiation of both moderator heavy water and rod bays light water should be pursued. AECL has already

embarked on a program to significantly reduce the flow of tritium to the rod bays by modest detritiation of the moderator. This has the additional benefit of reducing dose to NRU personnel during handling of the moderator heavy water. The proposed target for this is about 4 Ci/kg, requiring a feed-and-bleed of about 40 Mg/a D₂O. The program for regular detritiation of the heavy water at Darlington Tritium Removal Facility is being prepared and will be implemented after construction of a truck transfer facility in NRU. With the reactor moderator held at 4 Ci/kg, the rod bays can be detritiated to maintain the tritium concentration at 1.5 mCi/L by implementing a rod bays water feed-and-bleed program, sized for a manageable throughput of 600 Mg/a.

One difficulty of relying on continuous detritiation to reduce the tritium concentration is that it takes a long time to approach steady state. For this reason, a one-time replacement of the moderator and the rod bays with low-tritium water is a good way to reduce tritium concentration in the rod bays immediately. During the recent extended outage of NRU, the moderator was drained and replaced with low-tritium heavy water, bringing the source term down from 25 Ci/kg to about 2.5 Ci/kg (resulting in the initial modest downward trend for the rod bays tritium concentration). However, a similar replacement of the rod bays water is not possible because the material contents of the bays cannot be removed from their water shielding. A novel solution has been tested and will be implemented in the near future: The water will be replaced with cold fresh water by thermally-stratified displacement. This will result in the rod bays being immediately brought down to a tritium concentration of less than 1.5 mCi/kg without generating an excessive amount of tritiated light-water waste.

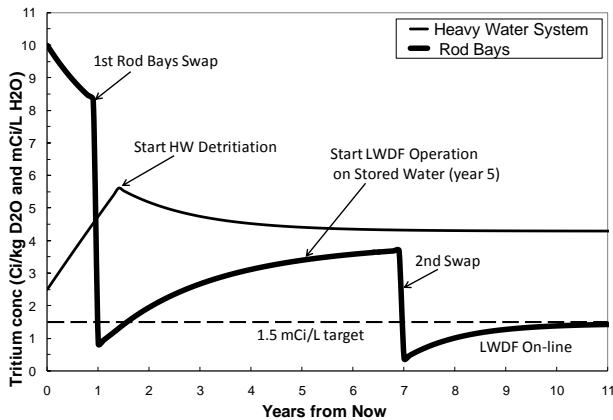


Fig. 1: Predicted Rod Bays Tritium Time-profile.

AECL is proceeding with setting up systems for continuous detritiation of both the NRU moderator and rod bays water. These will take some time: about 1.5 years for the moderator feed and bleed system and about 5 years to build a rod-bays Light Water Detritiation Facility

(LWDF). Figure 1 shows the modeled rod bays concentration in this case. It can be seen that the target tritium concentration is achieved in a reasonable time and without excessive tritium levels in the interim.

III. DETRITIATION TECHNOLOGY FOR LIGHT WATER

As is the case for pressurized water reactor tritiated water, the situation of this light-water detritiation issue has the following characteristics:

- Low tritium feed concentration.
- Comparatively large throughput.
- The need to achieve a substantial tritium enrichment to concentrate it into a relatively small volume.
- No need to produce a near-pure tritium product, such as is done in heavy water detritiation.
- If the treated water is to be discarded, then the process effluent must be stripped of tritium to a level that is environmentally acceptable, typically requiring $>10^5$ reduction in tritium concentration

One further point, particular to a heavy-water reactor situation: enriching the tritium also enriches the deuterium, producing a product that can be upgraded at a CANDU station and reused as makeup heavy water.

The CECE process is well suited to the light-water detritiation problem. This is primarily because of the magnitude of the separation factor and the low tritium concentrations in the process that do not raise tritium emissions, hazard, inventory and compatibility issues. The H-T separation factor in a hydrogen-water system operating at typical CECE conditions (60°C) is 5.3—compared with about 1.06 typical for water distillation. The relatively large separation factor means that a CECE process can achieve a large tritium separation between bottom product and overhead effluent streams in a relatively short catalyst bed (e.g., 10^8 -fold tritium separation in ~30 m of catalyst bed). The ability of the CECE process to strip the effluent to natural abundance tritium level has a particular advantage if the objective is to discharge the water to the environment.

The high separation factor also means that, relative to water distillation, the CECE process is physically small. For CECE light-water detritiation, the ratio of internal column flow to feed flow is typically 1.5. By contrast, the ratio of internal column flows to feed flow for water distillation is 15-25 and consequently, much larger columns are required.

IV. LIGHT-WATER DETRITIATION FACILITY (LWDF) DESIGN

Figure 2 shows a simplified flowsheet for the proposed rod bays LWDF. Tritiated rod bays water fed to the detritiation facility is split into a tritium-concentrated product stream and a near-tritium-free effluent hydrogen

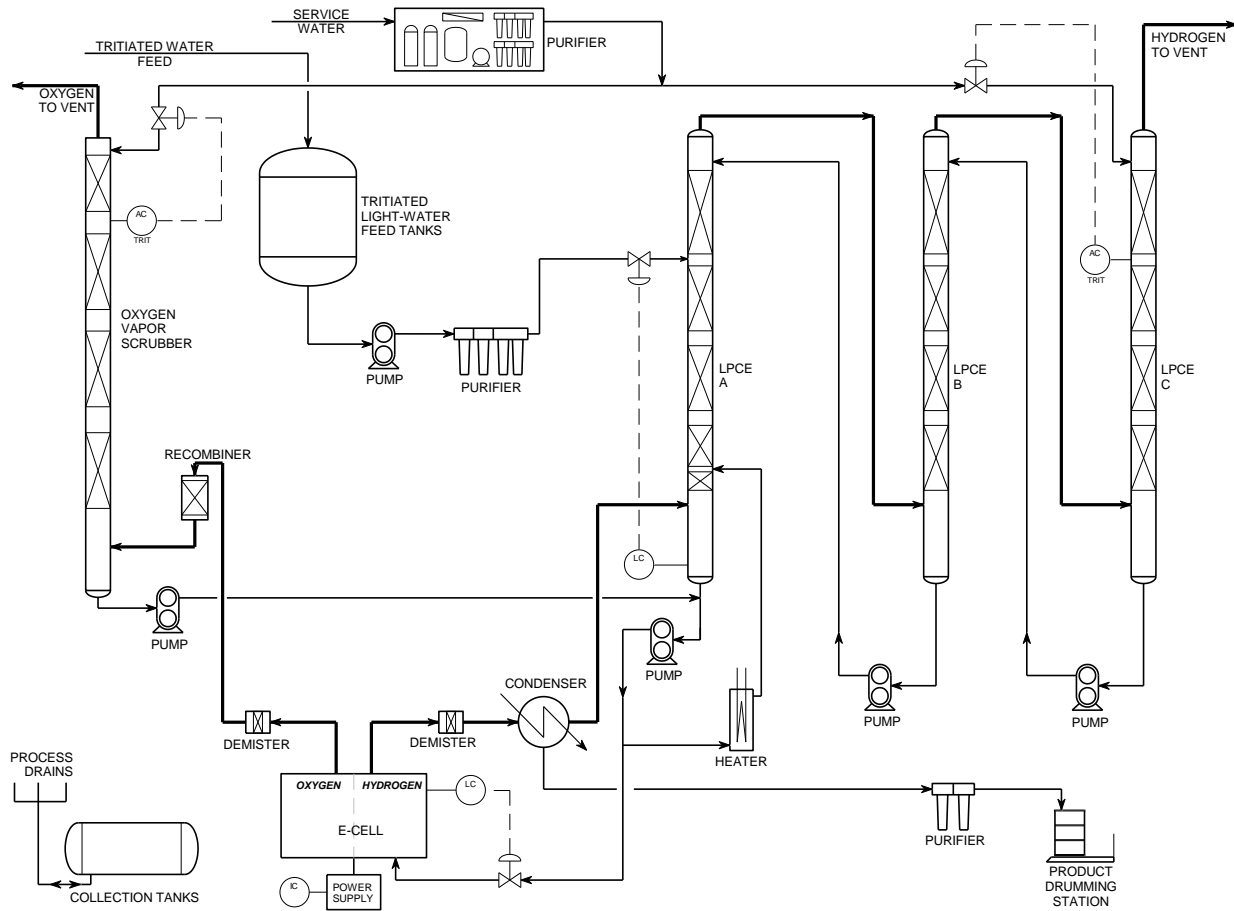


Fig. 2: LWDF Simplified Flowsheet

stream. The facility does not return detritiated water to the rod bays. Rod bays water is converted to hydrogen, stripped of tritium and vented. Deionized natural water is used for rod bays replacement water. Although the CECE process is quite capable of reducing tritium in the effluent hydrogen to below natural abundance levels (typically about 5 Bq/L at CRL), this maximizes the amount of catalyst required. In this proposed design, to rationalize catalyst requirements, hydrogen is discharged with a tritium content less than the equivalent of 2.7 nCi/L (<100 Bq/L) as water (this is less than 2% of the Canadian drinking water standard). The stripping section is fed with natural deionized water at a flow rate of 50% of the feed flow from the rod bays.

The LWDF is designed to be capable of a tritium enrichment of at least 1000 with a limit of 2 Ci/L at the point of highest concentration—in the electrolysis cells. This tritium concentration limit is chosen because of its licensing significance: systems containing tritium at more than 2 Ci/kg must be classified under Canadian regulations as Nuclear Class 3. Product water is drawn from electrolysis cell condensate, which is free of caustic

electrolyte and at only a slightly lower tritium concentration than the electrolyte. The draw-off is at a rate calculated to hold the tritium concentration in the electrolysis cells at 1.7 Ci/L—just below the regulatory limit. The tritium that was fed to the process from the rod bays, is thus removed in drums as downgraded heavy water, with a tritium concentration of 1.7 Ci/L and heavy water concentration of 10-90%. Initially about 15 drums per year would be produced, but after the stored rod bays water (from the early swap) is processed and the on-going detritiation brings the rod bays to the target concentration, the rate would quickly decrease to less than 2 drums per year.

The substantial oxygen stream from the electrolysis cells contains a significant amount of tritium—both as water vapour and as a hydrogen impurity. A catalytic hydrogen-oxygen recombiner converts trace hydrogen in the oxygen stream to water. An oxygen stream vapour scrubber, fed at the top with deionized natural water, scrubs tritium from the water vapour carried with the electrolytic oxygen effluent down to a tritium concentration of less than 2.7 nCi/L (<100 Bq/L).

The design of the LWDF is based on the output of a detailed process model developed at Chalk River Laboratories specifically for hydrogen/water exchange processes. The model has been used to size the following major equipment:

- 0.35 m diameter LPCE columns packed with AECL type 86-93 random catalyst, overall catalyst height of 30 m divided into 3 separate columns
 - 0.15 m diameter OVS column packed with structured mass transfer packing, overall packed height of 11 m
 - 40 L of AECL type 99-11 catalyst in a gas-phase recombiner
- Other major equipment sized independently includes:
- Two 4 m³ rods bays water storage tanks to ensure continuous supply
 - Three sets of water purification trains suitable for the bays water feed of 72 kg/h, 50 kg/h of normal water and up to 0.5 kg/h of product water.
 - Two tanks large enough to collect the total process inventory of electrolyte and water to facilitate maintenance activities.
 - 330 kA of electrolysis configured as one cell package, containing about 2 m³ of hydrogen gas and about 3 kCi of tritium—the majority of the plant inventory.

V. TECHNOLOGY READINESS

The CECE light-water detritiation process is based on the same technology used for CECE heavy water detritiation. CECE heavy water detritiation has been successfully demonstrated on a pilot scale at Chalk River (the CECEUD¹) and on a small production scale at NSSI², Houston. The lifetime and robustness of AECL's hydrogen-water exchange catalyst in an industrial environment has been demonstrated at the Prototype CIRCE Plant³; a 2.5-year demonstration of heavy water production by the CIRCE process at Air Liquide's site in Hamilton, Ontario. The IMET water electrolysis cell, marketed by Hydrogenics of Mississauga, is AECL's reference technology for the CECE process. IMET cells are a demonstrated industrial technology. IMET cell materials have been tested at Chalk River Laboratories for radiation resistance by exposing the IMET cell stack materials to high doses of gamma radiation. The tests have demonstrated the mechanical integrity of the cell materials at equivalent tritium exposure levels far greater than what would be experienced over the lifetime of the LWDF described here.

VI. CONCLUSIONS

The most practical solution to detritiate the NRU rod bays is to provide a storage system capable of holding at least one full volume of water coupled with a detritiation plant sized for 600 Mg/a throughput. This will quickly

reduce the rod bays tritium concentration from the current level of about 10 mCi/L and ultimately hold it below 1.5 mCi/L. A CECE light-water detritiation process conceptual design is presented. This once-through process discharges rod bays water as hydrogen at an equivalent tritium concentration of less than 2% of the Canadian drinking water standard and concentrates tritium in water about 1000-fold. The almost 10⁸ tritium separation is achieved in about 30 m of catalyst bed. The process has a maximum tritium concentration of <2 Ci/L and a tritium inventory of about 3 kCi.

The CECE process is well suited to the light-water detritiation problem. The relatively large H-T separation factor of 5.3 for hydrogen-water exchange makes it possible to achieve a large tritium separation top-to-bottom in a relatively short column. Tritium can be enriched 1000-fold without creating a significant tritium oxide hazard or challenging the tritium compatibility of electrolysis cells. A CECE light-water detritiation process is significantly smaller than a comparable water distillation process.

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