PNNL-39061

A Brief Review of Technologies for Separating Tritium from the Aqueous Effluent of Reprocessing Plants

February 2026

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Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

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operated by
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for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC05-76RL01830

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1.0 Introduction

1.1 Background

Tritium is a fission and activation product present in uranium oxide Used Nuclear Fuel (UNF). It is a relatively low energy beta emitter and poses relatively low radiological risk to humans and the environment. In an aqueous-based reprocessing plant, the tritium forms tritiated water when the UNF is dissolved in nitric acid and, unless otherwise mitigated, becomes dispersed and diluted in the light (non-radioactive) water throughout the plant. A small quantity of tritiated wastewater may be stabilized in a solid matrix and dispositioned as Low Level Waste (LLW). However, the larger quantity is currently discharged to the environment after removal of other contaminants because implementation of technologies to separate tritium for disposal have not historically been considered warranted because of its low radiological risk. That was the case for the late 20th century reprocessing plants in the United Kingdom, France, and Japan as considered by Arm (2025). In the U.S., environmental regulations limiting the discharge of tritium to the environment and the increasing viability of technologies for separating tritium will likely lead to a requirement of a future reprocessing plant to separate tritium for disposal.

Technologies at the front and backends of a reprocessing plant have received attention. At the front end, voloxidation is a technology that has received considerable attention in the U.S. national laboratories, particularly Oak Ridge National Laboratory. Voloxidation involves heating the UNF to between 450 and 600°C in an oxygen or nitrogen dioxide atmosphere that converts the uranium dioxide to a fine powder of uranium trioxide or triuranium octoxide (U_3O_8). The chemical change causes the fuel to break from the cladding and release volatile fission products, such as tritium, into the off-gas. The tritium can then be separated from the off-gas, stabilized and dispositioned as a relatively small volume.

Back-end technologies have received some attention in the U.S. but not as much as they have in Europe. These technologies include molecular sieves, distillation, and a collection of technologies identified as Combined Electrolysis and Catalytic Exchange (CECE). Advancements in back-end technologies have recently been spurred by fusion energy development and the Fukushima nuclear disaster in particular.

1.2 Scope

The scope of this letter report encompasses a review of technologies applicable to separating tritiated water from light and heavy water with a focus on wastewater arising from the reprocessing of UNF. Reports considering flowsheet options for tritium management are also reviewed.

1.3 Objectives

The main objective of this letter report is to support consideration of programmatic opportunities for the Materials Recovery and Waste Form Development (MRWFD) Campaign within the U.S. Department of Energy (DOE) Office of Nuclear Energy.

2.0 Flowsheet Studies

2.1 PUREX Adaptations

One issue with separating tritiated water from reprocessing operations wastewater is its dispersion throughout the flowsheet and its consequent low concentration. While there are established low-risk technologies for separating tritiated water (e.g., distillation), the operation becomes energy intensive given the volume and low concentration of tritium in the wastewater. Nonetheless, the PUREX flowsheet has evolved to facilitate the concentration of tritiated water and minimize its dispersion. Tritium control in the PUREX flowsheet is practical by the limited solubility of water in the PUREX solvent, tri-butyl phosphate (TBP) in a diluent such as dodecane (e.g., see Fujine et al. (1989) and Uchiyama et al. (1995)) and facilitated by the recycle of tritiated water. Figure 1 illustrates the concept of tritium control in the adapted PUREX flowsheet.

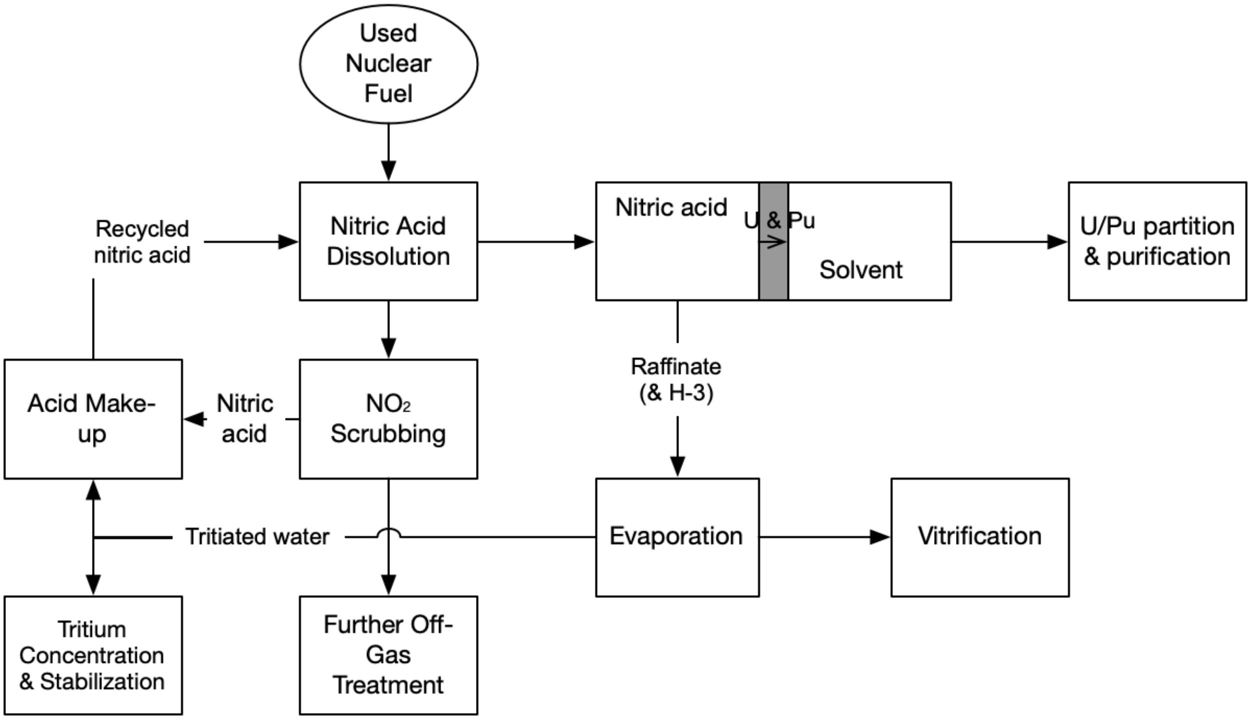

Figure 1. Concept of PUREX Adaptation for Tritium Control

With reference to Figure 1, UNF is dissolved in concentrated nitric acid before uranium and plutonium are separated from the minor actinides and fission products in a solvent extraction system. The separated uranium and plutonium are then partitioned and purified for fabrication into fuel. The solvent extraction raffinate containing the minor actinides, fission products, and most of the tritiated water are concentrated by evaporation and then vitrified for geologic disposal. The evaporated water, including tritiated water, is recycled for nitric acid make-up together with the nitric acid recovered from the dissolver off-gas by scrubbing for nitrogen dioxide gas. The recycled nitric acid is then used for dissolving UNF. A bleed of tritiated water is taken off for concentration, stabilization (e.g., grout), and disposal. Inevitably some tritiated water dissolves in the solvent and disperses through the flowsheet. Therefore, the flowsheet is said to be divided into high and low tritium sections.

This general concept was promulgated by Michels et al. (1978) and envisioned isotopic distillation as the technology for tritium concentration. These authors also point out that the release of tritiated water through the process ventilation system can also be significant but calculated it is still much less than the limit at the site boundary assuming process cell and vent improvements (e.g., driers). Project costs associated with process cell and vent improvements were calculated to be approximately the same as the tritium rectification and stabilization systems. Alternatively, a portion of the recycled tritiated water could be directly stabilized without concentration by rectification and was considered economically advantageous.

Henrich et al. (1980a) and Heinrich et al. (1980b) considered application of a tritium scrub to further reduce the quantity of tritiated water passing to the low tritium section by means of the solvent. Such a scrub could be achieved on the solvent containing the extracted uranium and plutonium using non-tritiated (i.e., fresh) nitric acid at a high (25:1) organic to aqueous phase ratio. Henrich et al. (1980a) postulate that any needed process cell and vent improvements then become limited to the high tritium section. As described by Arm (2025), tritium scrubs were implemented in the La Hague (France) reprocessing plants UP2-800 and UP3 and in the Rokkasho Reprocessing Plant in Japan. The solvent extraction system incorporating the tritium scrub is illustrated in Figure 2 below.

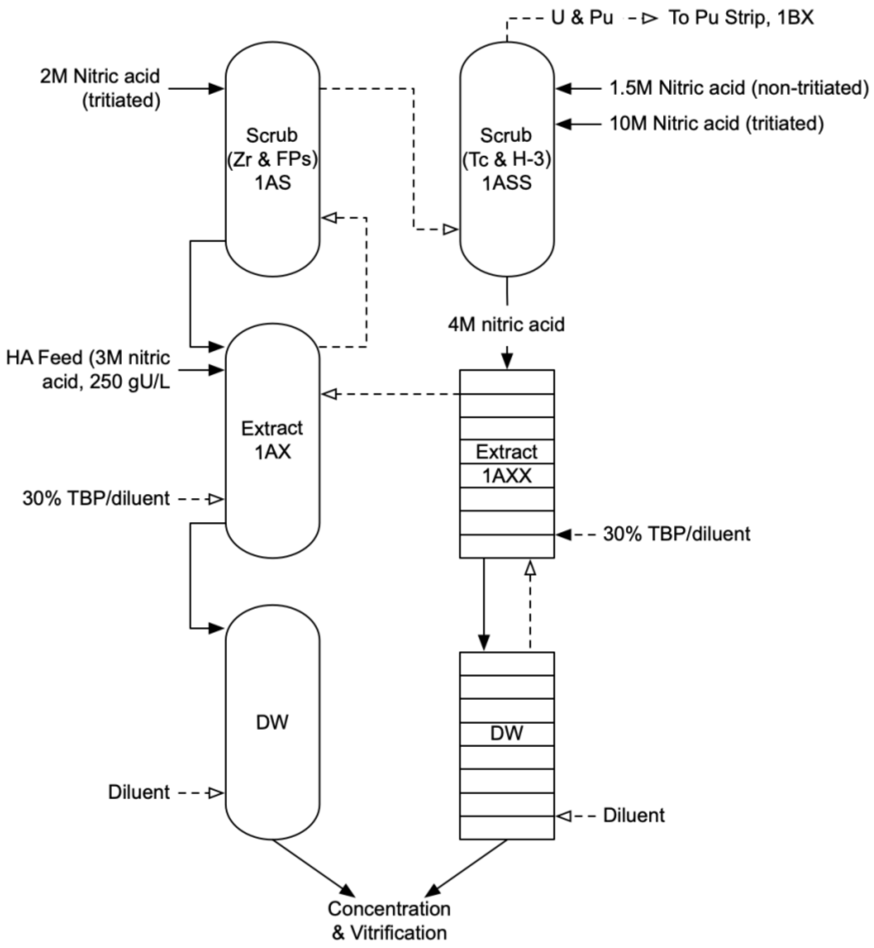

Figure 2. First Cycle PUREX Solvent Extraction Flowsheet Incorporating the Tritium Scrub

Here, the High Active (HA) feed is fed to the Extract pulsed column (1AX) where the uranium, plutonium and some fission products, including tritium, are extracted into the solvent. The solvent is scrubbed of fission products, notably zirconium, using 2M nitric acid in the 1AS pulsed column. The solvent is further scrubbed in column 1ASS to scrub tritium using 1.5M non-tritiated (i.e., fresh) nitric acid and technetium using 10M nitric acid. The scrubbed solvent then passes to systems that partition the uranium and plutonium and purifies them. The acid from the tritium scrub passes to the 1AXX extraction mixer-settlers where any residual uranium and plutonium are re-extracted and recycled to the 1AX column. The acid raffinates from the 1AX and 1AXX contactors are washed with diluent to remove residual TBP before concentration by evaporation and vitrification. Tritiated water is recycled to make up nitric acid in the manner illustrated in Figure 1. Any excess tritiated water is discharged to the ocean rather than stabilized although the author understands stabilization is being considered for a future reprocessing plant at La Hague. Purely by conjecture, the main reason for containing tritium to a high tritium section in the La Hague plants may be to facilitate potential tritium stabilization and disposal later if oceanic discharge were curtailed. Notably, the Thermal Oxide Reprocessing Plant at Sellafield, United Kingdom did not include any tritium control in its flowsheet, which perhaps adds credence to the motivation for the plants at La Hague.

In summary, adaptations to the PUREX flowsheet have successfully mitigated the dispersion of tritiated water and concentrated it in a single stream at the industrial scale.

2.2 Retention Flowsheet Alternatives Studies

One of the earliest alternatives studies identified was that of Burger and Scheele (1978). They considered various frontend technologies as alternatives to voloxidation but all were characterized by a high temperature similar to voloxidation. Water recycle was extensively studied and was considered by them to be viable in concentrating the tritium into a relatively small volume that could be stabilized and dispositioned. Burger and Scheele (1978) considered isotope separation would become a viable concept if combined with recycle.

Grimes et al. (1982) evaluated five options against a base case:

- The base case (option 1) does not retain any tritium and all tritium is discharged as tritiated water in excess light water to the atmosphere by evaporation.
- Option 2 involves concentrating the tritium by recycling 90% of the water. A side stream of the recycled water is dispositioned and 12% of the water and tritium is released to the atmosphere.
- Voloxidation was considered in option 3 with the recovered tritium dispositioned.
- Option 4 involved concentrating the tritium in the recycle of option 2 by various means and dispositioned.
- Like option 4, option 5 involved concentrating the tritium by various means from the non-recycled water and dispositioning it.
- Finally, option 6 involved isolating and concentrating the tritium in the head-end of a plant and dispositioning it.

This evaluation included both technical and economic considerations. Of the options, the third and fifth are of particular interest. Concerning option 3, the authors identify some engineering challenges with voloxidation, including:

- Reliability of the mechanisms for transferring fuel between the shear and voloxidizer and between the voloxidizer and dissolver.
- Dust confinement.
- Rotary voloxizider seal life.
- Removal of decay and reaction heat for process and cell temperature control.
- Isolation of voloxidizer process gases from shearing and dissolver equipment.

Grimes et al. (1982) considered the Combined Electrolysis and Catalytic Exchange (CECE) process for separating the tritium from the wastewater in option 5 (and 4). This technology is described later but received considerable attention in Europe where a pilot plant had been constructed. The main technical issue with the water recycle options was the much higher tritium concentrations in the recycled acid feed and the need for radiological controls. On the positive side, tritium was the only radionuclide present and so the equipment did not require containment with shielding. Between options 3 and 5, the latter had the lowest capital cost while the operating costs were comparable. That observation led the authors to conclude voloxidation did not appear economic.

Jubin and Spencer (2017) completed an evaluation of various tritium management scenarios that included water recycles and inclusion of tritium pretreatment (i.e., voloxidation) and tritiated water separation systems. They assumed an overall plant decontamination factor (DF) of 100 for tritium was required to limit radiological dose to the environment. Their work highlighted the importance of the vessel off-gas system and control of its dewpoint to achieving the overall plant DF. In all, the authors evaluated 19 scenarios covering different dewpoints for the dissolver off-gas, vessel off-gas, and melter off-gas systems and recycle configurations with tritium pretreatment and separation from aqueous effluent. Results for these scenarios in terms of volumes of tritium-contaminated wastes and recycle stream concentrations were presented. No scenario was recommended but they did recommend further work to demonstrate >99.5% removal of tritium in pretreating the UNF and performance of a technology for separation of tritiated water from aqueous effluent. The specific technology identified in the work was membrane-based, consistent with some experimental work ongoing at the time. That maturation of technology options on both the front and backends of a reprocessing plant was recommended is strategically important for the MRWFD Campaign.

3.0 Applicable Technologies

3.1 Technology Reviews

The review described by Khan et al. (2026) is mainly focused on proton exchange membrane electrolytic applications to fusion energy. However, the authors do compare various detritiation approaches in terms of separation factor and cost to show electrolysis offering the highest separation efficiency for a relatively moderate cost. Further, they suggest combining electrolysis with another technology such as catalytic exchange (to derive the CECE technology) provides a more cost-effective approach. The researchers recommend focusing on membrane durability, improving separation efficiency and reducing operational costs.

Zhao et al. (2025) have reviewed several technologies for separating tritiated water including: distillation, thermal diffusion, membranes, adsorption, catalytic exchange (vapor and liquid), and electrolysis. The researchers highlight CECE and liquid phase catalytic exchange as technologies that can offer high separation coefficients and large processing capacities. Although these technologies have begun to find industrial applications, the researchers identify a need to develop more durable catalysts and improve energy efficiency of electrolysis.

At the Hanford site in southeast Washington, radioactive liquid effluent arising from treatment of tank waste is treated to remove chemical and radiochemical constituents before discharge to a state-approved land disposal system. Tritium is not separated but is mitigated by its storage in slow-moving groundwater to allow extended time for radioactive decay before the water reaches the Columbia River site boundary. In this way, tritium in the state-approved land disposal system is isolated from the general environment and human contact until it has decayed to acceptable drinking water standards. Nonetheless, DOE is required to periodically evaluate alternative mitigation approaches. DOE-RL (2022) describes the latest evaluation. The authors evaluated five technologies: CECE, graphene oxide membranes, zeolite membranes, advanced water distillation, and nano filtration. Of these technologies, the authors identified CECE as the most promising. These technologies are described in the following sections.

3.2 Distillation

Tunaboylu and Paulovic (1991) presented a distillation facility to remove tritiated water from the wastewater arising from the proposed Wackersdorf Reprocessing Plant in Germany. At that time, the authors highlight distillation as the state-of-the-art technology with 40 facilities then operating for hydrogen isotope separation. Additionally, the authors consider it less hazardous than the electrolysis-based technologies because distillation avoids production of hydrogen gas. The disadvantage with distillation is the large equipment required; approximately 500 theoretical separation plates at reasonable vacuum and temperature. Therefore, distillation is performed in a few inter-connected columns. The authors project a thousand-fold volume reduction in tritiated waste by its enrichment. The number of theoretical plates is similar to that projected by Michels et al. (1978) who considered distillation of the high-tritium side wastewater of their flowsheet.

Although not directly applicable to reprocessing plant wastewater, Kalyanam and Sood (1988) illustrate the impact of cryogenic distillation and preceding electrolysis on distillation equipment size. They envision cryogenic distillation columns requiring 70 separation plates, about a tenth of those conceived by Tunaboylu and Paulovic (1991) in their columns at near-ambient pressure, at the expense of higher operating costs and more challenging hazards mitigation.

Chen et al. (2020) describe a water detritiation pilot facility based on distillation with a view to applying the concept to detritiating the wastewater from a fusion reactor. Their column had a packed section 12 m high and 0.26 m diameter. The packing was a phosphor bronze bilayer gauze corrugated structure. The researchers determined the Height Equivalent Theoretical Plate at 0.16 m for a feed of 13.5 MBq/L and flow rate of 25 kg/hour.

Busigen (2025) has presented an overview of distillation as it applies to light water detritiation, upgrading heavy water and heavy water detritiation in the context of CANDU nuclear power plants. All CANDU nuclear plants use water distillation for heavy water upgrading (the process of removing light water). For light water detritiation, Busigen (2025) reports a detritiation factor of 10,000 and an enrichment factor for 100 for a packed column height of 48.5 m and feed rate of 25 kg/hour containing 1 Ci/kg tritium.

In summary, water distillation is an industrially mature and effective technology for detritiating light water but involves large capital equipment.

3.3 Combined Electrolysis Catalytic Exchange

The Combined Electrolysis Catalytic Exchange (CECE) technology represents a combination of electrolytic and water-gas exchange separation approaches that individually offer tritium concentration but combined provide additional efficiency. The technology has received a lot of attention for a long period of time, particularly in Europe and Canada, for detritiating reprocessing plant wastewater and heavy water, respectively. The essential configuration of the CECE technology is illustrated in Figure 3. Here, tritiated wastewater is fed to an exchange column packed with a hydrophobic catalyst where it contacts hydrogen gas. Tritium in the gas preferentially exchanges with protium in the wastewater such that the water becomes enriched in tritium. The tritiated water flows to an electrolyzer where the non-tritiated water preferentially splits to hydrogen and oxygen that then flows up to the exchange column. The effect of the combined technologies is to produce a tritium enriched water product from the electrolyzer and detritiated hydrogen and oxygen gases.

Boniface et al. (2014) have provided a succinct description of the technology and provided capital and operating cost estimates for a plant to detritiate Fukushima wastewater. The authors point out that CECE costs scale linearly with throughput because they are dominated by the electrolyzer costs. They estimate capital and operating costs (in 2013 U.S.D) of \$345M and \$174M/year, respectively, for a plant treating 400 m^3/day.

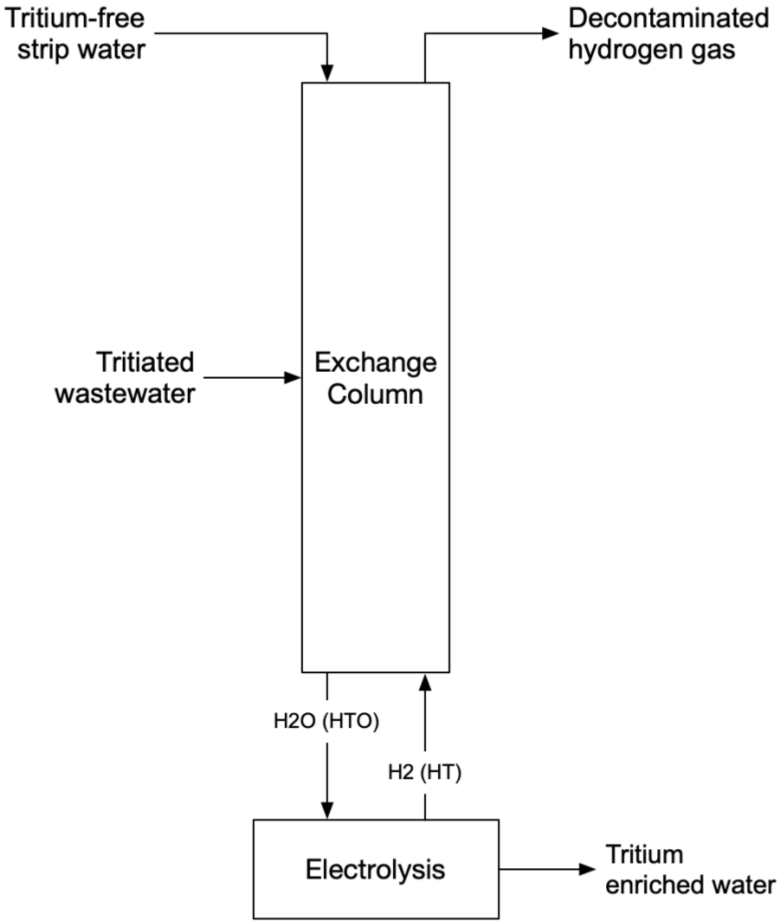

Figure 3. Basic Configuration of the CECE Technology

Phillips et al. (2013) selected the CECE technology for estimating the waste streams arising from a NUEX reprocessing system with a throughput of 800 MTHM/year. In their flowsheet, water was to be recycled with a bleed to account for freshwater additions. The bleed would be treated before disposal, including detritiation using the CECE technology which would concentrate the tritium to 200 Ci/L in a daily wastewater volume of 200 L. The feed to the detritiation plant was projected at 12,000 L/day or 500 L/hour with a tritium concentration of 0.3 Ci/L. Scaling the estimates of Boniface et al. (2014) based on the feed rate, the estimated escalated capital and operating costs are $11M and $5.5M per year, respectively, in 2020 U.S.D. The DOE-NE Systems Analysis & Integration Campaign (2021) have estimated an 800 MTHM/year reprocessing plant based on the French COEX technology with associated waste conditioning and storage to cost USD(2020)1.56B. Therefore, the cost of a detritiation facility based on the CECE technology is approximately 0.7% of the total of a reprocessing plant. No operations costs are provided by the DOE-NE Systems Analysis & Integration Campaign (2021) but the operating cost of a CECE plant is likely the same order of percentage of a reprocessing, waste conditioning and storage plant.

The following sections describe the various manifestations of the CECE technology but in summary the CECE technology appears to be considered the state-of-the-art and economic for detritiating wastewater.

3.3.1 ELectrolysis EXchange (ELEX)

The ELEX technology was developed at the Belgian Nuclear Research Center in the early 1980s with funding from the European Community (now European Union). The work of Bruggeman et al. (1980, 1982, 1984, 1985) and Geens et al. (1988) is probably the most extensively published of the CECE technology. Geens et al. (1988) presents the final report for the sponsor and describes a pilot plant consisting of a water electrolyzer and a trickle bed exchange column packed with a granular catalyst composed of platinum on charcoal mixed with PTFE and inert commercial rings (e.g., Raschig rings). It was designed for a throughput of 5 L/hour with a tritium concentration of 100 Ci/L, a decontamination and concentration factor of 100 and total tritium inventory of 1,000 Ci. These workers also evaluated various approaches to pretreat the liquid effluent and concluded adsorption with activated carbon and ion exchange were satisfactory. Geens et al. (1988) completed a cost estimate for adding the CECE process to a 150 MTHM/year plant and concluded its cost about 2% of that for the whole plant. Major areas of development leading to the pilot plant were catalyst formulation and electrolyzer configuration.

3.3.2 Veolia

Whitcomb et al. (2017) and Bonnet et al. (2015) have described one of the most recent manifestations of the CECE technology. Originally developed and demonstrated by Kurion to detritiate wastewater at Fukushima, the Modular Detritiation System (MDS) technology is now owned by Veolia. According to Veolia’s website, the prototype system was operated for over 1,000 hours in a 9-month period treating tritiated water at 1,300 L/day. Key to their technology are proprietary column internal materials that enable a much greater flow of gas than previously demonstrated. The column is 22-cm diameter and 9 m tall and achieves a 1000-fold volume reduction. The throughput of the MDS prototype is a tenth of that identified by Phillips et al. (2013) as required for an 800 MTHM/year reprocessing plant and would serve as a reasonable pilot plant. The feed tritium concentration to the MDS over the test period is not explicit. Included in the system is a metal hydride bed that immobilizes the separated tritium for storage.

3.3.3 Others

In Japan, Isomura et al. (1988) constructed a small pilot plant with a throughput of 3.6 L/day and demonstrated a tritium concentration factor of 10,000. Their catalyst was 0.3 to 0.6 wt% platinum in porous divinylbenzene copolymer. The electrolyzer contained a solid polymer of a NAFION membrane doped with platinum. Their tests showed the importance of minimizing dissolved metals and suspended solids and maintaining a neutral pH to maintain the performance of the membrane.

The Canadian nuclear industry has been detritiating the heavy water used in CANDU reactors for several decades using cryogenic distillation following electrolysis. More recently, the CECE technology has been explored and adopted as a more economic alternative. Miller et al. (2002) have described the initial tests of the technology using a pilot plant with a feed rate of 17 L/day and a tritium concentration of 10 Ci/L. Neither the catalyst nor the electrolyzer membrane were identified although the former was described as a ‘wet-proofed, isotope exchange catalyst’ that implies it is proprietary.

3.4 Molecular Sieves

Bhave et al. (2016a, 2016b, 2017) investigated the performance of two advanced zeolite membranes to concentrate tritiated water with funding from the MRWFD Campaign. The membranes included cation-exchanged silicoalumino phosphate molecular sieves and Linde Type A (LTA). Non-tritiated water preferentially passes through the LTA membrane, which provided the better performance. Based on experimental results, the authors projected needing a total membrane area of 8,153 m^2 of 17 stages to treat a flow rate of 60 L/hour containing tritium at a concentration of 1 Ci/L. They compare that to the dehydration of alcohols where LTA membranes with >1,000 m^2 are reported. However, the flow rate here is approximately an order of magnitude lower than that projected by Phillips et al. (2013) at 500 L/hour for an 800 MTHM/year reprocessing plant, which suggests a membrane area of 68,000 m^2 would be required. Their techno-economic analysis projected operating costs were an order of magnitude less for the membrane system. Capital costs appear not to be directly comparable. For the 60 L/day membrane-based facility, the capital cost was estimated at $5.1M. Capital costs for a CECE-based facility processing 600 L/hour (similar to that projected by Phillips et al. (2013)) was $3.4M, which is the same order as that projected at $10M using the data from Boniface et al. (2014). That comparison suggests the CECE-based facility would be less expensive than a membrane-based facility.

Sevigny et al. (2015) investigated the performance of graphene oxide membranes to concentrate tritiated water also with funding from the MRWFD Campaign. The workers observed some separation of tritiated water given the permeation of light water was greater. They recommended improving the membranes by including a hydrophilic coating to allow direct contact with water and provide additional strength. However, their cost analysis indicated a membrane-based facility would be significantly greater than one based on CECE. The work appears to have been discontinued.

Jeppson et al. (2000) presented proof-of-principle tests with a proprietary but commercially available ion exchange resin (likely sulfonated polystyrene/ divinylbenzene) loaded in 2 m tall columns that preferentially loaded tritiated water as water of hydration. They loaded the resin with aluminum ion that carries with it significant waters of hydration and then processed treated Hanford evaporator wastewater to show the tritiated water preferentially displaced the light water. The tritiated water was then desorbed by heating the resin at ~150°C after removing the light water at lower temperatures. Quite low concentrations of tritium were tested. No further work appears to have been done to mature this technology or to estimate the size of a plant.

In summary, approaches using molecular sieves and membranes are feasible but are likely more expensive than other technologies.

4.0 Conclusions and Recommendations

4.1 Conclusions

This work has shown there to be viable and technically mature technologies to detritiate the wastewater from reprocessing plants. The CECE technology, in particular, appears to be the state-of-the-art and has received considerable attention in Europe and North America. Nonetheless, further optimization of the technology should be pursued to leverage advances in its constituent technologies, electrolysis and catalytic exchange. These opportunities are briefly discussed below.

4.2 Recommendations

Arguably the CECE technology has matured to a significant level with industrial investment. However, the most recent developments have focused on applications other than treating reprocessing plant wastewater, such as treating wastewaters associated with Fukushima and nuclear power plants (light and heavy water moderated). Therefore, development of the technology tested for treating reprocessing plant wastewater in the 1980s accounting for materials advancements since then is warranted.

The electrolysis cells are the major capital and operating costs drivers. Therefore, any improvement in electrolyzer efficiency will benefit the technology in terms of its cost. Improvements may focus on electrode and membrane materials and general advances in the electrolysis technology.

Whereas the capacity of the electrolyzers and diameter of the catalytic exchange column is driven by the total flow of water, the height of the column is driven by the concentration of tritium. The packing is key to minimizing the height in ensuring intimate contact between the water and hydrogen gas and a fast exchange of hydrogen isotopes. Few details are published about the packing used by researchers and at least some are proprietary. Furthermore, there is a balance between the catalyst design and operating conditions. A low column temperature minimizes the strip water flow by maximizing the separation factor for tritium. On the other hand, a higher temperature operation uses less catalyst but requires more electrolysis. Column packing development while considering operating conditions is warranted to optimize the system for detritiating reprocessing plant wastewater.

A reprocessing plant's wastewater will require treatment to remove trace radiochemical and chemical constituents to environmental release standards. The order of such treatment steps would require optimization but detritiation has historically been considered the final step. Therefore, a relatively clean feed to a CECE system would be expected. Nonetheless, tests to understand the robustness of the CECE system to feed impurities would be desirable.

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