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2014 EVALUATION OF TRITIUM REMOVAL AND MITIGATION TECHNOLOGIES FOR WASTEWATER TREATMENT, DOE/RL-2014-10, REVISION 0, AND HANFORD FEDERAL FACILITY AGREEMENT AND CONSENT ORDER (TRI-PARTY AGREEMENT) MILESTONE M-026-07C

The purpose of this letter is to transmit the 2014 Evaluation of Tritium Removal and Mitigation Technologies for Wastewater Treatment, DOE/RL-2014-10, Revision 0, in accordance with Tri-Party Agreement Milestone M-026-07C due March 31, 2014.

If you have any question, please contact me or your staff may contact Al Farabee, of my staff, on (509) 376-8089.

Sincerely,

J C Perry FOR

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DOE/RL-2014-10,
Revision 0

2014 Evaluation of Tritium Removal and Mitigation Technologies for Wastewater Treatment

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

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DOE/RL-2014-10,
Revision 0

2014 Evaluation of Tritium Removal and Mitigation Technologies for Wastewater Treatment

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Executive Summary

Since 1995, a state-approved land disposal site (SALDS) has received tritium contaminated effluents from the Hanford Site Effluent Treatment Facility (ETF). Tritium in this effluent is mitigated by storage in slow moving groundwater to allow extended time for decay before the water reaches the site boundary. By this method, tritium in the SALDS is isolated from the general environment and human contact until it has decayed to acceptable levels.

This report contains the 2014 update evaluation of alternative tritium mitigation techniques to control tritium in liquid effluents and groundwater at the Hanford Site. A thorough literature review was completed, and updated information is provided on state-of-the-art technologies for control of tritium in wastewaters. This report was prepared to satisfy the Tri-Party Agreement (Ecology et al., 1989, *Hanford Federal Facility Agreement and Consent Order*) Milestone M-026-07C. Tritium separation and isolation technologies are evaluated periodically to determine their feasibility for implementation to control Hanford Site liquid effluents and groundwaters to meet the 40 CFR 141.66[1] drinking water maximum contaminant level for tritium of 20,000 pCi/L and/or DOE Order 5400.5[2] as low as reasonably achievable policy.

Since the 2009 evaluation, there have been a number of developments related to tritium separation and control with potential application in mitigating tritium contaminated wastewater. These are primarily focused in incremental improvements to the existing technologies in use such as improved catalysts, membranes, and electrolysis technologies.

Continuing development efforts for tritium separations processes are primarily to support the International Thermonuclear Experimental Reactor program, the nuclear power industry, and the production of radiochemicals. While these applications are significantly

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[1] 40 CFR 141, "National Primary Drinking Water Regulations," *Code of Federal Regulations*. Available at: http://www.gpo.gov/fdsys/pkg/CFR-2010-title40-vol22/xml/CFR-2010-title40-vol22-part141.xml.

[2] DOE Order 5400.5 Chg 2, 1993, *Radiation Protection of the Public and the Environment*, U.S. Department of Energy, Washington, D.C. Available at: https://www.directives.doe.gov/directives/archive-directives/5400.05-BOrder-c2/view.

different than the Hanford application, the technology could potentially be adapted for Hanford wastewater treatment.

Separations based processes to reduce tritium levels below the drinking water standard have not been demonstrated for the scale and conditions required for treating Hanford wastewater. Available cost information indicates that treatment costs for such processes will be substantially higher than for discharge to the SALDS or other typical pump and treat projects at Hanford. Actual mitigation projects for groundwater with very low tritium contamination similar to that found at Hanford have focused mainly on controlling migration and on evaporation for dispersion in the atmosphere.

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Terms

AECL	Atomic Energy of Canada, Limited
ANL	Argonne National Laboratory
BLIP	Brookhaven Linear Accelerator Isotope Producer
CECE	combined electrolysis catalytic exchange
CERCLA	*Comprehensive Environmental Response, Compensation, and Liability Act of 1980*
DF	decontamination factor
DOE	U. S. Department of Energy
DWS	drinking water standard
EPRI	Electric Power Research Institute
ETF	Effluent Treatment Facility
FY	fiscal year
GE	GE-Hitachi Nuclear Energy
ITER	International Thermonuclear Experimental Reactor
LPCE	liquid phase catalytic exchange
MDS	Modular Detritiation System
MNA	monitored natural attenuation
MVR	mechanical vapor recompression
NTP	number of theoretical plates
PNPI	Petersburg Nuclear Physics Institute
RCRA	*Resource Conservation and Recovery Act of 1976*
ROD	record of decision
SALDS	State-Approved Land Disposal Site
SRS	Savannah River Site
TRS	Tritium Resin Separation
VLB	Viscous Liquid Barrier
WDS	Water Detritiation System
WTRF	Wolsong Tritium Removal Facility

Elements and Compounds

Hydrogen Isotopes	Atomic Mass
H (1H) protium	1.008
D (2H) deuterium	2.014
T (3H) tritium	3.017

Pure tritium with a mass of 1.03 grams is equivalent to 10,000 Ci

D2	deuterium molecule
D2O	deuterium oxide (heavy water)
DT	tritiated deuterium
DTO	tritiated deuterium oxide
H2	hydrogen molecule
H2O	water (light water-protium oxide or protinated water)
HD	deuterated hydrogen
HDO	deuterated water
HT	tritiated hydrogen
HTO, T2O	tritiated water
T2	tritium molecule

Abbreviations

g	gas
l	liquid
Kd	distribution coefficient
p(H2O)	partial pressure of H2O
p(HTO)	partial pressure of HTO

1 Introduction

Tritium was generated as a byproduct in reactor fuel at the Hanford Site by the U.S. Defense Program in nuclear reactor operations from 1944 to 1989. The bulk of this tritium was released to the ground from fuel reprocessing facilities in the 200 Area Central Plateau in the form of tritiated waste in process condensate. Releases to the ground have greatly decreased since the last fuel was processed through the fuel separations plant in 1989. Tritium inventories remain in the Hanford Site groundwater and in the underground waste storage tanks, 100 KW basin water, and waters stored at the 200 East Area Liquid Effluent Retention Facility. Concentrations of tritium in Hanford groundwater continue to decline as a result of natural decay and dispersion. Tritium decays with a 12.3 year half-life into helium.

The annual Hanford Site groundwater monitoring report describes the monitoring results for *Resource Conservation and Recovery Act of 1976* treatment, storage, and disposal units and, *Comprehensive Environmental Response, Compensation, and Liability Act of 1980* (CERCLA) groundwater operable units and to meet the requirements of the *Atomic Energy Act of 1954*. The U.S. Department of Energy (DOE) operates an extensive groundwater monitoring program on the Hanford Site, collecting thousands of samples from hundreds of well each year. The latest report issued in August of 2013 reported the monitoring results for 2012 (DOE/RL-2013-22, *Hanford Site Groundwater Monitoring Report for 2012*). This reports shows that concentrations of tritium are declining in many of the River Corridor and Central Plateau wells as the plumes attenuate naturally by radioactive decay and dispersion.

Since 1995, a state-approved land disposal site (SALDS) has received effluents from the Hanford Site Effluent Treatment Facility (ETF) that are essentially free of all contaminants except tritium.
The majority of tritium discharged to the SALDS, during the period 2009 to 2013, came from the K Basin wastewater, 242-A Evaporator condensate, and treated groundwater streams. Discharge to the SALDS allows natural radioactive decay to reduce tritium content substantially before the wastewater enters the Columbia River.

The numerical groundwater flow and contaminant transport model of the SALDS tritium plume was last updated in fiscal year (FY) 2011 (SGW-53569, *Results of Tritium Tracking and Groundwater Monitoring at the Hanford Site 200 Area State Approved Land Disposal Site, Fiscal Year 2012*). The model included tritium migration and fate predictions based on both the latest calibration of the groundwater model and the latest information regarding the forecast operation of the 200 West Pump and Treat. The modeling results showed that some locations along the northern margin of the 200 West Area are expected to exhibit measurable concentrations of SALDS-derived tritium by the year 2030, although the model indicates that concentrations would be below the drinking water standard (DWS) of 20,000 pCi/L.
The eastern end of the tritium plume is also predicted to migrate to the south toward the 200 West Pump and Treat extraction wells by 2030. The maximum tritium concentration in the SALDS proximal wells in 2009 to 2013 was 180,000 pCi/L. To date, tritium from the SALDS has not been detected in any of the tritium-tracking wells, although Well 299-W6-6 has seen increased tritium due to reinjection of effluent from the 200 West Pump and Treat.

Currently, the chosen alternative for handling tritium in the ETF effluent is to discharge this water to the subsurface and allow the tritium to decay into non-radioactive helium before it reaches the Columbia River. Although it would be desirable to have an alternative process that could actively separate tritium from water in the environment, the much larger cost to benefit ratio makes this approach unrealistic. Therefore, at this time, discharge to the subsurface is the only cost effective method to handle tritium in feed to the ETF (Ecology, 2000, *State Waste Discharge Permit Number ST 4500*).

Under the 200-ZP-1 record of decision (ROD) (EPA et al., 2008, *Record of Decision Hanford 200 Area 200-ZP-1 Superfund Site Benton County, Washington*), which specifies operation of the 200 West Pump and Treat as the selected remedy for removal of contaminants from groundwater, the preferred alternative for tritium treatment is monitored natural attenuation (MNA).

In 1991, the DOE Richland Operations Office (Site Technology Coordination Group) issued a notice of the need to reduce tritium concentrations in Hanford Site wastewaters from 2 to 3 million pCi/L to less than 20,000 pCi /L (DOE/RL-98-01, Hanford Science and *Technology Needs Statements, 1999*). *Fiscal Year 1999 Waste Tank Science Need* (RL-WT047-S) was issued that called for identification of viable processes for reducing tritium concentrations in Hanford Site wastewaters.

The current report is one in a series of the following documents concerning tritium mitigation technologies:

- DOE/RL-94-77, *Tritiated Wastewater Treatment and Disposal Evaluation for 1994*, provided an initial evaluation of tritium treatment and disposal options.
- DOE/RL-95-68, *Tritiated Wastewater Treatment and Disposal Evaluation for 1995.*
- DOE/RL-97-54, *Evaluation of Tritium Removal and Mitigation Technologies for Hanford Site Wastewaters.*
- DOE/RL-99-42, *1999 Evaluation of Tritium Removal and Mitigation Technologies for Wastewater Treatment.*
- DOE/RL-2001-33, *2001 Evaluation of Tritium Removal and Mitigation Technologies for Waste Water Treatment.*
- DOE/RL-2004-11, *2004 Evaluation of Tritium Removal and Mitigation Technologies for Wastewater Treatment.*
- DOE/RL-2009-18, *2009 Evaluation of Tritium Removal and Mitigation Technologies for Wastewater Treatment.*

The current report provides an update of developments in the area of tritium mitigation technology since the 2009 update. The earlier reports should be consulted for additional background information that is not repeated herein.

2 Summary

Separations based processes to reduce tritium levels in Hanford groundwater below the DWS of 20,000 pCi/L have not been demonstrated for the scale and conditions required for treating Hanford Site wastewater. Available cost data indicate that treatment costs for such processes will be substantially higher than for discharge to the SALDS or other typical pump and treat projects at the Hanford Site. The total cost for processing wastewater through the ETF is less than $1/gal. Cost for SALDS disposal is estimated to be less than $0.01/gal.

To determine if new technologies may be technically and economically feasible to reduce tritium in the Hanford ETF outfall, a literature search was conducted to update information in earlier reviews. Separation processes were evaluated for application to ETF effluent. Other potentially applicable methods for mitigation of tritium contaminated wastewater or groundwater at the Hanford Site were also considered. The conclusions from this review are that S&GWR tritium separations technology has advanced, but nothing has materially changed that would make alternate technologies preferable to SALDS discharge for handling the ETF effluent.

The ETF outfall is characterized as a high flow (5 to 28 Mgal/yr) low tritium concentration wastewater. Peak tritium concentrations of 450,000 to 550,000 ρCi/L occurred periodically in 2009 through 2013 during treatment of K-Basin water and the 242-A Evaporator process condensate. This is down from the previous five year period where the peak tritium concentration was 2 to 3 million pCi/L during 2004 through 2008. To meet the current DWS (20,000 ρCi/L), a nominal 100-fold reduction (decontamination factor [DF] = 100) in tritium concentration is required.

Currently, there are no technologies available that can selectively remove the trace amounts of tritium from the ETF outfall in a single simple step. Rather, complex multistage separations processes are required because only a small degree of tritium separation can be achieved in a single stage. Conventional heavy water production methods (e.g., distillation, combined electrolysis and catalytic exchange [CECE]) are highly energy intensive, and the high costs cannot be justified for recovering a small amount of tritium from the ETF outfall.

Development work since the last update report has continued in support of heavy water reactors and nuclear fusion reactors which entail tritium removal from lower volume, higher contamination level streams. Most of the work is focused on incremental improvements to the existing technologies in use, such as separations processes based on water distillation, CECE, improved catalysts, and membranes. Atomic Energy of Canada Limited (AECL) has successfully demonstrated the CECE and cryogenic distillation process for heavy water detritiation in semi-industrial scale (TalkNuclear, 2011, *CNA Visits AECL's Chalk River Laboratories PART TWO*; Rodriquez, 2013, *Tritium Production Analysis and Management Strategies for a Fluoride-salt-cooled High-temperature Test Reactor (FHTR)*; Ionita, 2013, *Experimental Tritium Removal Facility an example/support for national/international collaboration*).

Kurion introduced a new tritium removal technology in October 2013 (Kurion, 2013a, *Kurion Introduces Tritium Removal Technology to Limit Release of Radionuclides into Environment*). The industrial process of removing tritium from water has historically focused on cleaning highly contaminated heavy water for recycling back into nuclear reactors. However, this technology is prohibitively expensive for use with light water reactors. Kurion claims that their new Modular Detritiation System (MDS) (Kurion, 2013b, *Modular Detritiation System™ (MDS™)*) builds upon proven heavy water solutions and makes advances in throughput and efficiency where the tritium removal occurs. Kurion suggests that they are using a derivative of the technology used in Canada to remove tritium from heavy water that moderates the Canada Deuterium Uranium reactors used there. However, their claim has yet to be proven on a large

scale. The Secretary of DOE is promoting Kurion to help with the Fukushima Plant (EXSKF, 2013, *U.S. Energy Secretary Ernest Moniz to Japanese Government: "Ratify the Convention on Supplementary Compensation for Nuclear Damage, and We Will Send Kurion to Help TEPCO"*).

An alternative to tritium removal is tritium mitigation, where the wastewater is discharged in a location that takes advantage of the short half-life of tritium (about 12.3 years). One approach for mitigation of tritium in wastewater is to store it underground in a stagnant or slow moving aquifer where it will be isolated from the general environment and human consumption for an extended period of time. This approach has the advantages of low cost and low energy consumption, provided a suitable storage location is available. This method is being used at the Hanford SALDS facility where groundwater migration rates are closely monitored to prevent environmental contamination of the biosphere.

Other tritium mitigation methods have been implemented at several sites with wastewater and groundwater with trace tritium contamination. These methods include implementing actions to restrict or alter groundwater movement by pumping or forming barriers, or by evaporation for air dispersion (thermal evaporation or "phytoremediation," i.e., the use of plants to uptake and evaporate groundwater). These "last ditch" efforts are not required for the Hanford SALDS facility, which provides isolation from the environment while tritium concentrations decay to relatively low levels.

Under the 200-ZP-1 ROD (EPA et al. 2008), the preferred alternative for tritium treatment at the 200 West Pump and Treat is MNA. This alternative is preferred because there is currently no viable treatment technology to remove tritium from the groundwater. However, the half-life of tritium is sufficiently short, so the tritium will decay below the cleanup standard before it leaves the industrial land-use zone.

3 Removal and Mitigation Technologies for Tritium in Wastewaters

The majority of the tritium removal technologies have been developed for wastewater containing tritium at levels higher than expected in Hanford Site wastewater or observed in Hanford Site groundwater. However, as presented in Table 1, several technologies (shaded in blue) are applicable to Hanford Site wastewaters having less than 1.0E-05 Ci/L of tritium. This report discusses those technologies applicable to Hanford Site wastewaters. The status designations for these technologies have not changed from the 2009 report as reflected in the last column of the table. Other technologies listed cannot reasonably be applied on the scale required to address Hanford Site tritium contamination. The soil column discharge concept (see Section 3.3.3) has been implemented via the use of the SALDS for treated effluent disposal.

3.1 Industrial Hydrogen-Isotope Separation Technologies

Hydrogen isotope separation technologies include processes that separate deuterated water (HDO) and deuterium oxide (D_2O) from water (light water-protium oxide or protinated water [H_2O]) and/or tritiated water (HTO) from HDO and D_2O. None of these processes are used on a large commercial scale for separating very low concentrations of tritium from light water to meet the drinking water concentration. Processes discussed in this section would require some work to be adapted to the Hanford wastewater treatment requirements, but this is considered to be a moderate extrapolation from past successful applications of the processes.

3.1.1 Water Distillation

Water distillation is simple and based on equipment used in various chemical process industries. There are few rotating components, and the units are sealed. Isotope separation by water distillation is based on the small differences in vapor pressure between water species containing different hydrogen isotopes. Water distillation for separation of HDO and D_2O from H_2O is a safe and well-established process that has been used on an industrial scale at commercial heavy water nuclear reactors for many years in the United States, Canada, and Europe. Typical feed rates to commercial heavy water plants are ~1 gallons per minute (gpm). Water distillation has generally not been used to remove traces of HTO from large volumes of wastewater (IAEA, 2012, *Efficient Water Management in Water Cooled Reactors*) such as the ETF outfall.

3.1.1.1 Water Distillation – Process Description

Water distillation separation is based on the relative volatility of HTO and H_2O. At 60°C, the H_2O vapor pressure is about 1.056 times that of HTO. Thus, the equilibrium liquid mole fraction of HTO is 1.056 higher than the gas phase mole fraction. To reduce overall energy consumption and cooling water requirements, mechanical vapor recompression (MVR) is utilized to heat the reboiler. The overhead vapor stream is compressed and it condenses in the reboiler. The condensing energy of the overhead steam is used for boiling the bottoms product in the reboiler. By this method, approximately 90 percent of the energy used for standard distillation is conserved. Even with MVR, water distillation is a high energy consuming method (IAEA, 2012).

As presented in the previous report (DOE/RL-2009-18), for the Hanford application a DF of about 100 is needed, so the HTO concentration in the product needs to be reduced by 99 percent compared to the feed. To achieve this separation, several hundred distillation stages (theoretical plates) are required.
The number of theoretical plates (NTP) varies with the reflux ratio (ratio of recycled condensate to net product). A higher reflux ratio is more energy intensive, but reduces the NTP. The relationship between reflux ratio and NTP required for the upper (stripping section) of the distillation column is illustrated by Figure 1.

Table 1. Summary of Tritium Removal and Mitigation Technologies

Technology	Year Report Prepared						
	1994	1995	1997	1999	2001	2004	2009/ 2014
Water Distillation	D, h		D, h	D, h	D, h	D, h	D, h
Gaseous Diffusion	D, h						
Laser Isotope Separation	T, h	T, h	T, h				
Electrolysis	D, h	D, h	D, h				
Combined Electrolysis and Catalytic Exchange (CECE)	D, h	D, h T, l	D, h T, l	D, h T, l	D, h T, l	D, h T, l	D, h T, l
Combined Electrolysis Catalytic Exchange with Vapor Phase Catalytic Exchange					D, h		
Membrane Separation Process	T, l	T, l	T, l				
Cryogenic Distillation	D, h	D, h	D, h	D, h	D, h		
Bithermal Catalytic Exchange		D, h T, l		D, h T, l	D, h T, l	D, h T, l	D, h T, l
Isotopic Exchange, Air Sparge		T, l					
Finely Divided Nickel Catalyst		O					
Separation by Metanetix Inc.		O					
Substituted Naphthalene		O					
Crown Ether Complexes		O					
Girdler-Sulfide Process			D, h	D, h	D, h	D, h	D, h
Palladium Membrane Reactor							D, h
GE Integrated Systems							D, h
Liquid Phase Catalytic Exchange with Solid Oxide Electrolyte			D, h	D, h T, l	D, h T, l		
Liquid Phase Catalytic Exchange with High-Temperature Steam Electrolysis (Hot Elly)			D, h				
Sulfur Resin Ion Exchange			O				
Metal Hydride Exchange			T, h				
Soil Column Discharge	D, l, h		D, l, h	D, l, h	D, l, h	D, l, h	D, l, h

Table 1. Summary of Tritium Removal and Mitigation Technologies

Technology	Year Report Prepared						
	1994	1995	1997	1999	2001	2004	2009/ 2014
Barrier Formation			O	D, l, h	D, l, h	D, l, h	D, l, h
Air Sparging			T, l				
Dual-Temperature Liquid-Phase Catalytic Exchange				D, h			
Tritium Resin Separation Process				T, l	T, l	T, l	T, l
Kinetic-Isotope Effect for Concentrating Tritium				T, l	T, l		
Pumping and Recharging				D, l	D, l	D, l	D, l
Phytoremediation					D, l	D, l	D, l
Evaporation						D, l	D, l

Maturity:

D = Demonstrated or developed technology that has been successfully applied in the field

T = Testing or theoretical stage of development

O = Observation indicates a potential process needing funding to continue

Applicability:

l = Technology is applicable to larger wastewater volumes having lower levels of tritium (less than 1.0E-05 Ci/L)

h = Technology is applicable to smaller wastewater volumes having higher levels of tritium (greater than 1.0E-05 Ci/L)

As shown by the diagram, a reflux ratio of ~30 is reasonably optimum for balancing required NTP with required reflux flow. The NTP is not reduced much as the reflux ratio increases above 30, and the required NTP begins to increase rapidly if the reflux ratio is reduced much below 30.

By countercurrent contacting of ascending vapor and descending liquid, tritiated water is depleted from the overhead product and enriched in the lower (rectifying) section of the column. Liquid from the column bottom flows to the reboiler. A small fraction of the water in the reboiler (10 percent of the feed) is removed and sent to a second distillation column for further concentration. Two-stage distillation reduces the overall size of the equipment.

To evaluate feasibility of water distillation, a rough estimate of column sizing and power was made based on the assumptions shown in Table 2.

Based on the stated assumptions, it is estimated in the previous report for the Hanford application (DOE/RL-2009-18) that 230 theoretical plates are required for the primary column. The height equivalent of a theoretical plate for this application is about 4 in. using bronze gauze packing. This requires a packing height of 76 ft and a total column height of approximately 100 ft for the first column.

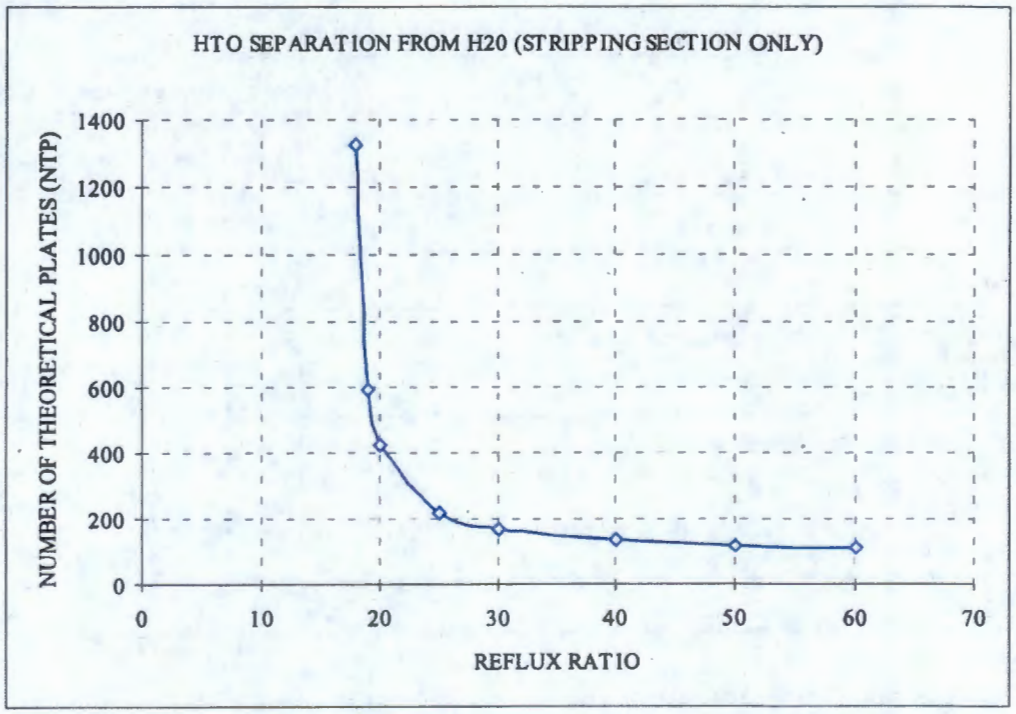

Figure 1. Water Distillation – Theoretical Plates Versus Reflux Ratio for a Decontamination Factor of 100

Table 2. Column Sizing and Power Rough Estimate Assumptions

Number of Parallel Process Trains	10
Wastewater Feed Rate	100 gpm (0.38 m3/min) Total 10 gpm (.04 m3/min) Per Train
Primary Column Parameters	
Product Water Tritium Concentration	0.001 times feed concentration
Reboiler Concentrate	Reboiler bottoms concentrated to 10 times feed concentration, 0.1 times feed volume
Liquid Reflux	30 times feed rate
Distillation Temperature	60°C Nominal (varies from top to bottom due to pressure variation in column)
Secondary Column Parameters	
Product Water Tritium Concentration	0.1 times concentration of feed transferred from primary column reboiler
Reboiler Concentrate	Reboiler bottoms concentrated to 100 times primary column reboiler concentration or 1,000 times wastewater feed concentration. Net volume is 0.001 times wastewater feed volume

Liquid Reflux	30 times feed transferred from primary column reboiler
Distillation Temperature	60°C Nominal (varies from top to bottom due to pressure variation in column)

The high reflux ratio results in high liquid and vapor flows (about 30 times the feed rate) and, consequently, large column diameters. If the feed is split into 10 parallel trains, each primary column would be approximately 24 ft in diameter. Each secondary column is estimated to require about 170 theoretical stages with dimensions of approximately 8 ft in diameter and 80 ft tall.

Based on the assumed conditions stated above, each primary and secondary distillation column would require 4,800 and 430 horsepower respectively for vapor recompression. Total compression horsepower for the 10 distillation trains is about 52,000 horsepower or 40 megawatts (MW) of power. Assuming a nominal power cost of $0.04 per kilowatt hour and operation 70 percent of the time, power is estimated to cost $10 million/year or ~$0.27/gal for recompression energy alone. While this is lower than earlier estimates, it is clear that the total costs would be more than an order of magnitude higher than costs for SALDS disposal or typical pump and treat projects (see Section 3.4 for additional information on relative costs between mitigation options).

3.1.1.2 Water Distillation – Recent Developments

GE-Hitachi Nuclear Energy (GE) has recently implemented water distillation for tritium removal of a relatively small waste stream at a radiochemical processing facility in the United Kingdom. GE has also proposed water distillation for treatment of wastewater at commercial nuclear power plants. A key feature of the GE process is the use of vapor recompression to reduce energy consumption and cooling water demand. This is a high energy consuming method (IAEA, 2012).

3.1.1.3 Water Distillation – Application at the Hanford Site

The limitation of water distillation lies in the large quantities of water that must be evaporated. Because the separation factor is small, the internal flows in the boilers and the condenser must be very high in comparison with the feed flow. Consequently, achieving the required minimum DF factor of 100, the equipment would be large and the energy consumption would be high. To reduce the overall energy consumption, MVR could be utilized to heat the reboiler. Water distillation has not been used as a detritiation process to treat large volumes (106,000 gal/day) of wastewater with low tritium activity as needed for the Hanford application (Candu et al., 2013, *Requirements for Tritium Removal Technologies*).

The technology is expected to work; however, the capital and operating costs are expected to be high. The process would require a large number (approximately 10) of 24 ft diameter primary towers each with 4,800 horsepower compressors plus additional smaller second stage columns and compressors. The capital and operating costs for water distillation would be more than an order of magnitude higher than costs for SALDS disposal or typical pump and treat projects (see Section 3.5 for additional information on relative costs between mitigation options). Therefore, use of this technology cannot be justified to recover the trace amount of tritium in the Hanford ETF outfall.

3.1.2 Combined Electrolysis Catalytic Exchange (CECE)

As presented in the previous report (DOE/RL-2009-18), CECE is one of several processes based on use of the hydrogen/water exchange equilibrium reaction (Equation 1) that favors formation of liquid HTO when liquid H_2O is contacted with tritiated hydrogen (HT) gas.

$$HT\ (g) + H_2O\ (l) \leftrightarrow HTO\ (l) + H_2\ (g) \qquad (1)$$

The CECE process has a high isotopic separation factor at near ambient temperature and pressure operating conditions. A catalyst is required for the reaction to proceed at an appreciable rate, and development of improved hydrophobic catalysts in recent years has been important to commercial success of the process.

The CECE process requires electrolysis of all feed water plus some deionized water used for stripping (approximately 1.4 times the feed flow is electrolyzed).

$$2H_2O_{(l)} \rightarrow 2H_{2(g)} + O_{2(g)} \qquad E_0 = 1.229\ V \qquad (2)$$

$$2HTO_{(l)} \rightarrow 2HT_{(g)} + O_{2(g)} \qquad E_0 > 1.229\ V \qquad (3)$$

The electrolysis separates tritiated water into elemental hydrogen (H_2), tritiated hydrogen (HT), and oxygen (O_2) gases. H_2O is more easily electrolyzed than HTO, so that H_2O is depleted from the liquid causing the HTO concentration in the electrolyzer liquid to increase. The CECE process is energy intensive because of the requirement to electrolyze 1.4 times the feed water.

A variation on the CECE process uses a palladium membrane reactor to separate elemental hydrogen from water to provide the required elemental hydrogen feed to the catalytic exchange unit. While not a direct method for producing hydrogen isotopes, the palladium membrane reactor generates a crude product for further purification by hydrogen isotope separation processes. The palladium membrane reactor technology is expected to provide some incremental cost savings for selected tritium separation processes. An isotopic separation process supported by the palladium membrane reactor has not been used at near the scale required at Hanford. These processes are considered to be excessively costly and need further development. This process also requires handling of large volumes of toxic and/or potentially explosive gas mixtures (e.g., carbon monoxide and hydrogen) resulting in additional safety hazards (IAEA, 2012).

3.1.2.1 CECE - Process Description

A schematic drawing of a CECE process is shown in Figure 2. The process consists of countercurrent gas/liquid exchange columns packed with catalyst beds, an electrolysis cell, and a hydrogen/oxygen recombiner (omitted if hydrogen co-production is desired). A platinum based hydrophobic solid catalyst is used. Tritiated water is added mid-column. As the water flows down the column, the tritiated hydrogen is transferred from the rising gas stream to the descending liquid stream by Equation 1.

The rising hydrogen gas stream is partially depleted in tritium in the bottom section of the column. In the top section, clean water further reduces the tritium content of the rising hydrogen, resulting in a hydrogen stream exiting the top that is nearly exhausted of tritium.

The combined water stream (feed plus added clean water) exits the bottom of the column to an electrolysis cell where it is electrolyzed to oxygen, hydrogen, and tritiated hydrogen gases by Equations 2 and 3. The enriched tritium stream can be taken from the bottom of the column as tritiated water or tritiated hydrogen gas depending on the desired form of the product.

3.1.2.2 CECE – Recent Developments

The CECE process has been the subject of active development work in recent years. The work includes catalyst development and testing, improvements to electrolytic cells, optimization of system and component designs, and industrial prototype construction and operation. Most of this work is aimed at tritium separation for heavy water reactors.

AECL has been a leader in developing advanced technologies for detritiation applications. AECL has successfully demonstrated the CECE and cryogenic distillation process for heavy water upgrading and

detritiation in semi-industrial scale (Boniface et al., 2010, *A Small Closed-Cycle Combined Electrolysis and Catalytic Exchange Test System for Water Detritiation*; INFO-0796, *Evaluation of Facilities Handling Tritium Part of the Tritium Studies Project*; Rodriquez, 2013; Ionita, 2013). These developments have been achieved through the experience gained from tritium handling and engineering in facilities. Though the AECL CECE technology has been demonstrated, an extensive research program has been ongoing to advance this technology further, particularly in electrolysis cell materials (Ana et al., 2013, *Construction and Commissioning of a Hydrogen Cryogenic Distillation System for Tritium Recovery at ICIT Rm. Vlacea*; Suppiah et al., 2010, *Tritium and Technology Developments for its Management A Canadian Perspective*).

The experimental industrial plant for hydrogen isotope separation on the basis of the CECE process has been operating safely and reliably for 15 years at the Petersburg Nuclear Physics Institute (PNPI) (Alekseev et al., 2011, "Fifteen Years of Operation of CECE Experimental Industrial Plant in PNPI"). Large scale studies of hydrogen isotope separation have been ongoing at the PNPI plant since 1995. Through plant operation, the studies have shown a high efficiency of isotope separation by the CECE process, and the process is considered to have promise for the industrial use, particularly water purification from tritium. The plant processing output has nearly doubled since startup and is now at nearly 9.0 kg/day (about 2.3 gal/day) and has demonstrated a tritium DF of 1,000 when operated with heavy water (a more difficult separation than with light water). Multiple operating modes and conditions have been tested.

The Water Detritiation System (WDS) test facility called TRENTA that applies the CECE process has been installed and is currently operating at the Tritium Laboratory Karlsruhe in Germany. The WDS is required for the International Thermonuclear Experimental Reactor (ITER) tritium plant in order to process tritiated water which is accumulated within the plant. The WDS plant was designed and commissioned to perform various experimental campaigns to investigate and optimize operational parameters to achieve the highest DFs, in view of different operation scenarios. The plant combines two well-established methods for the CECE process, which appear to be the most suitable process for water detritiation and tritium recovery. These methods involve the electrolysis of tritiated water, followed by a liquid phase catalytic exchange (LPCE) column. The WDS has demonstrated that the CECE process is capable of processing tritiated waste in larger amounts (greater than 2,000 gal/year) with tritium content in the range of about 0.01 Ci/L. However successful, the throughput is significantly short of the throughput required for Hanford Site water. Besides the ongoing research and development work, the current status of the TRENTA facility provides the option to utilize WDS for processing tritiated water (Michling et al., 2013, "Water Detritiation Processing of JET Purified Waste Water using the TRENTA Facility at Tritium Laboratory Karlsruhe").

Detritiation research has seen activity with the successful design, construction, and commissioning of the Wolsong Tritium Removal Facility (WTRF) in Korea. The WTRF uses an LPCE and cryogenic distillation process (Han, 2013, *Operating Experience and Effect of Wolsong Tritium Removal Facility*). Romania has constructed a pilot plant that uses similar detritiation technology as the WTRF in Korea. Both facilities are designed for low volume (<5 gal/hour) and very high tritium concentration (over 8 Ci/L). The Romania pilot plant has main goals of developing the heavy water detritiation technology and monitoring the behavior of various materials and equipment working with tritium (Stefan, 2009, *Detritiation in Romania*; Zamfirache et al., 2013, *Research Activities Related to Water Detritiation at ICIT Rm Valcea*).

Kurion introduced a new, patent pending tritium removal technology in October of 2013 (Kurion, 2013a). The Kurion MDS is based on advancements in the CECE process tailored for light water detritiation. Kurion claims that the unique electrolyzer and column design provides the highest possible throughput

per area and per energy. Kurion states that the system can easily be expanded to process 100,000 gal/day (Kurion, 2013b); however, their system has not been demonstrated on a large scale.

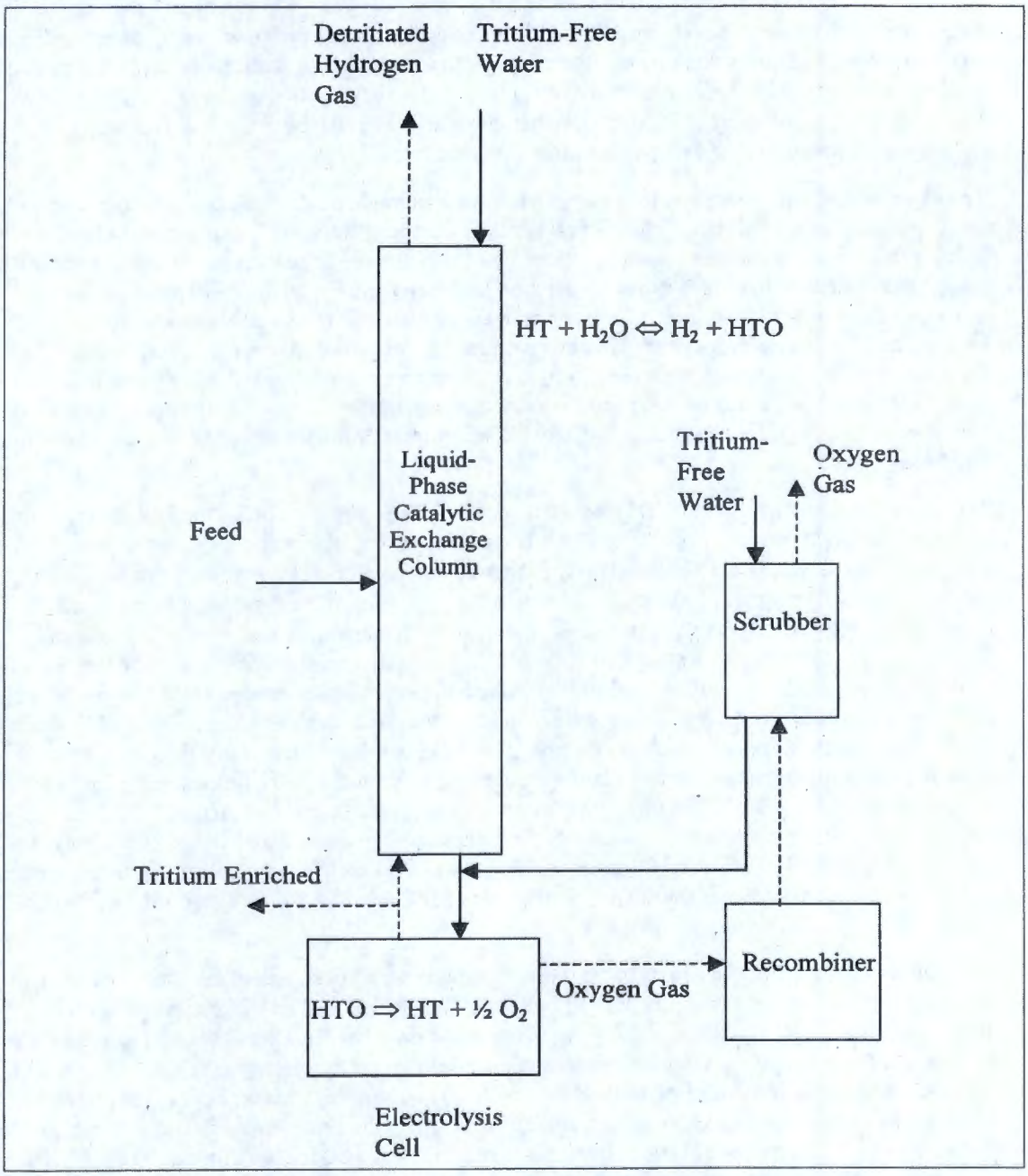

Figure 2. Combined Electrolysis Catalytic Exchange Process

3.1.2.3 CECE - Application at the Hanford Site

The CECE process has only been demonstrated on a small scale (32 gal/day) (Candu et al., 2013). No commercial facility able to treat the volume needed for the Hanford application is available.

Although there have been recent developments with process improvements, electrical power remains the major cost. The CECE process requires electrolysis of all feed water plus some deionized water used for stripping (approximately 1.4 times the feed flow is electrolyzed). About 164 MW of power to the electrolyzers is required to support a nominal 100 gpm wastewater feed rate. Power losses for conversion of incoming power to the low voltage direct current required for electrolysis and other power uses increase required power further above this value. This process requires a costly hydrophobic catalyst, considerable cost for electrolytic cells, and handling of hydrogen gas at near-atmospheric pressures for the separation. Confinement systems are required for the concentrated tritium product for protection of workers, the environment, and public. Detailed site specific cost estimates are not available and testing has not been performed with Hanford specific waste compositions.

Feed for this process needs to be water with low levels of organic and inorganic contaminants. Water that has been processed at the ETF is likely to work well as a feed because this wastewater is essentially free of contaminants other than tritium. Full characterization and possibly testing work would be needed to determine if additional pretreatment is required for specific candidate waste streams.

A tritium enriched waste stream will be produced in addition to tritium depleted water or hydrogen. This can be in the form of HT in hydrogen gas from the electrolytic cell or water with elevated tritium compared to the feed water. HT could be loaded on a metal as a hydride, or tritiated water could be dispositioned as a grouted waste form. This stream is small compared to the wastewater feed. Cost of dispositioning the waste will depend upon the method and could be significant.

A recent cost estimate by AECL indicated a treatment cost of approximately $1.9/gal (IAEA, 2012). Since energy cost for CECE tritium separation is approximately four times the cost for conventional distillation using MVR, the CECE process is not expected to be competitive for treating Hanford tritiated water.

3.1.3 Bithermal Hydrogen/Water Process

The bithermal hydrogen/water process is based on the same hydrogen/water exchange reaction as the CECE process (see Section 3.1.2) and may be able to use similar catalysts. However, it does not require electrolysis of the feed water but instead relies on a recycled stream of hydrogen coupled with dual temperature separations columns.

3.1.3.1 Bithermal Hydrogen/Water – Process Description

As presented in previous evaluations, the process consists of cold-stripping and cold-enriching columns and hot-enriching and hot-stripping columns stacked in a vertical orientation with hydrogen gas flowing upward countercurrent to the aqueous streams, as shown in Figure 3. Tritiated water to be treated is introduced between the cold-stripping and cold-enriching columns. Three conditions are important to maximizing separation factors: 1) use of an active hydrophobic catalyst, 2) temperature control to enhance the stripping and enriching conditions, and 3) high pressure. Hydrophobic catalysts are used, similar to the CECE process. However, some catalysts developed for CECE are not suitable because their upper temperature limit is about 100°C, which is lower than the optimum temperature for the bithermal process.

In the upper "cold stripper" section, non-tritiated water is used to absorb tritium from the circulating hydrogen. The resulting hydrogen gas, essentially free of tritium, is recirculated to the hot-stripping column to remove tritium from the wastewater to be discharged. The tritium-rich product stream is withdrawn from between the cold and hot enrichment columns. The columns are operated at near 49 atmospheres pressure to achieve maximum separation factors. The hot enrichment and stripping

column sections are operated at about 443°K (170°C), and the cold-stripping and cold-enrichment column sections are operated at about 323°K (50°C).

3.1.3.2 Bithermal Hydrogen/Water – Recent Developments

The bithermal hydrogen/water process uses the same chemistry and similar catalysts to those used for the CECE process. Therefore, much of the development work on the CECE process is directly or indirectly applicable to bithermal hydrogen/water. The technology was successfully demonstrated by AECL at the prototype Combined Industrial Reforming and Catalytic Exchange demonstration project at Hamilton, Ontario, Canada (Candu et al., 2013).

3.1.3.3 Bithermal Hydrogen/Water Process – Application at the Hanford Site

The bithermal hydrogen/water process has not been operated on a large industrial scale. The separation columns, catalyst beds, and internal stream flows are much larger than the CECE, and the operating costs are high (Candu et al., 2013).

Existing applications for the bithermal hydrogen/water process are for treating heavy water; however, it appears feasible to adapt the technology for treatment of Hanford Site wastewater. This process does not require electrolysis of the feed water to change phases of the feed stream, but it operates with large volumes of hydrogen gas at high pressure, heating to moderately high temperatures and significantly higher recirculation flows compared to the CECE process. The process is expected to be capable of reducing tritium concentrations from levels typical of Hanford wastewaters to less than the DWS of 20,000 ρCi/L for the depleted discharge stream while producing a small volume tritium-rich stream of >0.02 Ci/L. AECL has stated that this process can be designed to process 80 to 130 gpm with no obvious difficulty.

Feed water for this process needs to contain low levels of organic and inorganic contaminants. Water that has been processed at the ETF is likely to work well as a feed because this wastewater is essentially free of contaminants other than tritium. Full characterization and possibly testing work would be needed to determine if additional pretreatment is required for specific candidate waste streams.

Concerns with this process include: 1) containment of tritiated water and tritiated hydrogen gas under high pressure, 2) safety with the use of high-pressure hydrogen gas in the process, and 3) processing has not been used on a large industrial scale. The process is much more sensitive to control of the process flows than the CECE process. Because electrolysis of all the feed is not required, power costs are expected to be lower than for the CECE process. However, the separations columns, catalyst beds, and internal stream flows are much larger. As in the case of the CECE process, a method must be provided to disposition the concentrated tritiated water stream. As stated in the previous Hanford report (DOE/RL-2009-18), the process was evaluated to be only slightly more costly than the CECE process for the 20 gpm scenario evaluated. Lower DFs required at Hanford compared to the Savannah River Site (SRS) could tend to favor the bithermal hydrogen/water process because its costs are more sensitive to required separation efficiency. New studies indicate that the total treatment costs (e.g., capital, utilities, and labor) for this process are expected to be similar to the costs for the CECE process with the lowest cost option depending on capacity, operating duration, power cost, and other site specific factors (IAEA, 2012).

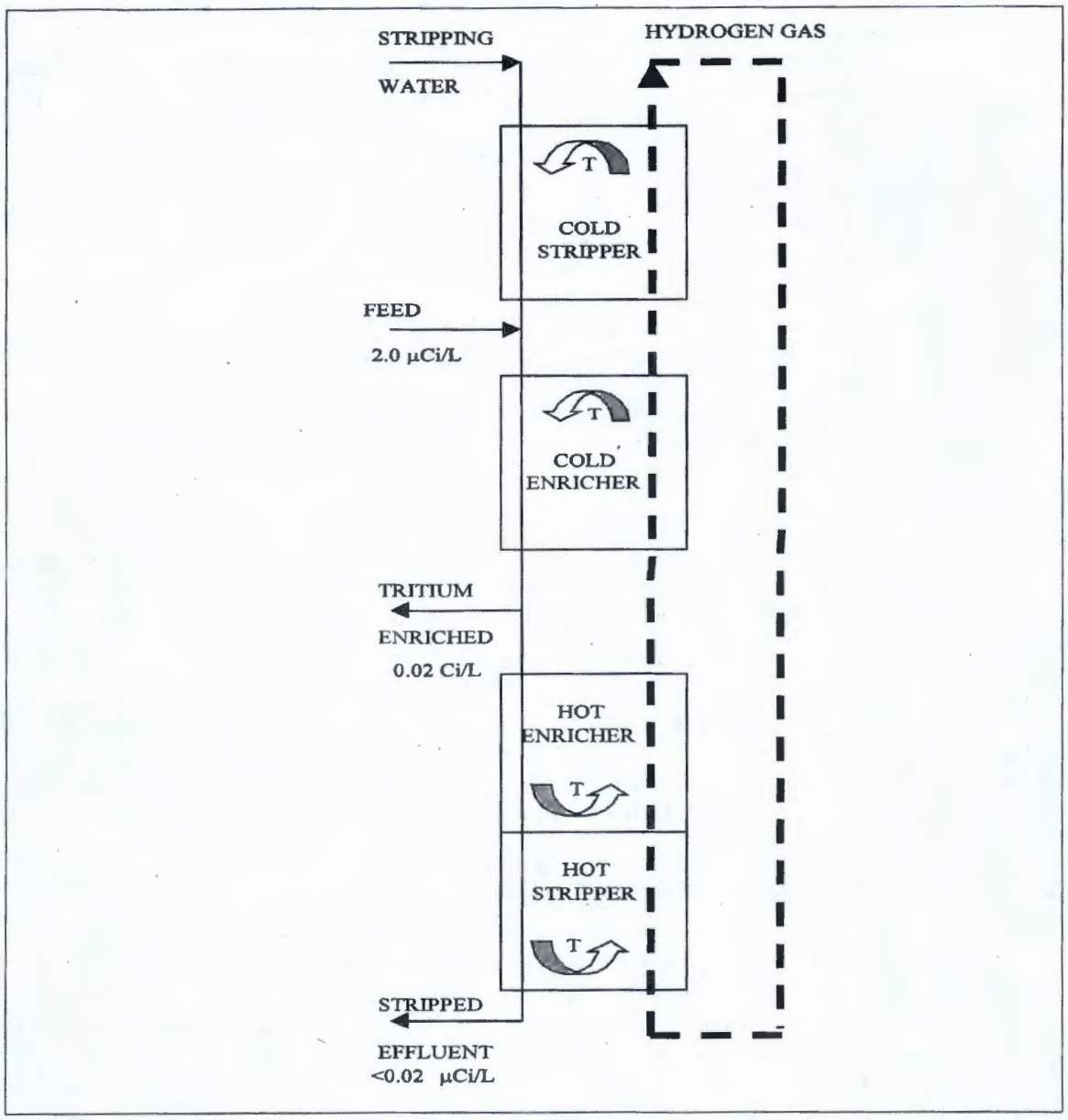

Figure 3. Bithermal Hydrogen/Water Process

3.1.4 GE Integrated Systems

As presented in the previous report (DOE/RL-2009-18), GE has developed several integrated system concepts for processing tritium contaminated wastes. These systems are designed to reduce environmental emissions and, in some cases, recover tritium for beneficial use.

The initial integrated process was installed at the GE manufacturing site located near Cardiff, United Kingdom, and resulted in a large reduction in tritium discharges to the environment. Building on this experience, GE has developed additional separations technology and integrated system concepts and is marketing them commercially. The three applications have the following significant features:

- Cardiff, United Kingdom Process. This process supports a facility that has been producing tritium-labeled chemicals since the 1940s. Thermal oxidation is performed first to convert the tritium component of organic material to tritiated water. The tritiated water is then processed through a water distillation system that concentrates the tritium in the bottoms. The concentrated tritiated water is mixed with carbon monoxide gas and fed to a palladium membrane reactor where the hydrogen isotopes are split from the water and separated from reaction products. The mixed hydrogen isotopes containing about 2,000 parts per million tritium are fed to a thermal diffusion system where tritium is separated from the other hydrogen isotopes, producing a concentrated product that is >99 percent tritium. Product tritium is recycled for beneficial use within the facility.

- Heavy Water. For heavy water decontamination, the proposed GE system uses liquid phase catalytic exchange (hydrogen/water) as the initial process step. The method for splitting elemental hydrogen isotopes from the feed water is not specified in the reference but would presumably be either electrolysis or the palladium membrane reactor depending on site specific conditions. The tritium rich stream from the catalytic exchange reactor is fed to gaseous diffusion, then thermal diffusion produces a high purity tritium product.

- ITER Fusion Reactor. To support tritiated water cleanup and tritium recycling for the ITER, the GE process uses water distillation as the initial step for processing. Bottoms from the water distillation system are transferred to liquid phase catalytic exchange. The tritium enriched stream from liquid phase catalytic exchange is processed by a gaseous diffusion cascade to produce a high concentration tritium product and a deuterium product.

GE has also evaluated systems for light water reactors (Busigin and Bonnett, 2010, *Light Water Reactor Detritiation Developments*; IAEA-TECDOC-1650, *Good Practices in Heavy Water Reactor Operation*). A recently completed study recommended a water distillation system for cleanup of tritiated wastewater at a commercial light water reactor (DOE/RL-2009-18). MVR technology may be beneficially used to reduce energy and cooling water consumption for this application (see Section 3.1.1 for additional discussion of distillation and MVR).

3.1.4.1 GE Integrated Systems – Application at the Hanford Site

The entry of GE into the tritium waste processing market is of interest because they bring an array of technology and expertise that can be brought to bear on tritium abatement issues. However, the specific tritium processes and systems they are involved with are at a very small scale compared to the Hanford wastewater volumes. While the proposed process concepts may provide some incremental improvement in performance and cost, they do not represent a major breakthrough that would substantially reduce cost for the Hanford application.

3.2 Tritium Adsorption Bed Separation

The tritium concentration in water effluents can be reduced to acceptable discharge levels by processing the water through an adsorption bed (DOE/RL-2009-18; Tanaka, 2011, "Water Vapor Adsorption Properties of Honeycomb-Type Zeolites for Tritium Removal Systems;" Wajima, 2011, "Adsorption Characteristics of Water Vapor on Zeolitic Materials for Honeycomb-Type Adsorbent"). The sorbent based Tritium Resin Separation (TRS) process developed by Molecular Separations Incorporated was discussed in earlier evaluation reports. In this process, a proprietary solid sorbent material is used. The sorbent has hydration sites that are selective for tritiated water (T_2O, HTO) over protinated (H_2O) water. Tritiated water is selectively adsorbed onto the sorbent as it contacts the contaminated water, and the sorbent is periodically regenerated by heating. Refer to evaluations in prior reports for a more detailed process description.

The sorbent based process was evaluated by the Electrical Power Research Institute (EPRI), including bench scale process testing. Major findings of the EPRI Assessment Test Program are summarized as follows:

- The TRS process selectively removes tritiated water from solution by a hydration mechanism. Removal efficiency rates range from 0 to 39 percent in beaker and column tests. The highest removal rates were seen in using an inorganic exchange resin.
- At this point the TRS process has not been shown to be an economically viable process technology. The technology can be characterized as having extremely short runs, long drying cycles, and relatively low tritium removal efficiency per cycle.
- Testing confirmed the requirement of complete removal of ionic and organic contaminants prior to the process.
- The testing of organic exchange resin resulted in a major failure due to rapid degradation of the media due to drying temperature.
- Operation of the test columns as a "fluidized bed" resulted in a major loss in the tritium DF and did not allow for the determination of the final tritium removal capacity for the media.

Based upon these findings, the TRS process has not been demonstrated to be suitable for removing tritium from Hanford wastewaters where tritium removal efficiency of 99 percent is needed, and the sorbent bed must be regenerable. A previous evaluation (DOE/RL-2004-11) also concluded that the treatment cost would be excessive even if the process was made to work. As a result of these findings, the TRS process is not viable for Hanford tritiated water treatment at this time.

3.3 Other Tritium Mitigation Technologies for Wastewaters

There are several concepts for delaying movement of tritium contaminated groundwater plumes, thereby maximizing the time before contaminated groundwater reaches site boundaries. These concepts are based upon the fact that tritium decays with a half-life of 12.3 years. Other mitigation methods involve evaporation or incineration of the tritiated water with releases directly to the atmosphere.

3.3.1 Pump and Recharge

The pump and recharge concept extracts tritium contaminated water from the ground and recharges it at a location where the movement of groundwater will take longer for the contaminated groundwater to reach site boundaries. Treatment to remove contaminants other than tritium may be performed prior to recharge. In some cases, this treatment approach is combined with methods to minimize the natural recharge to the aquifer in order to minimize the total volume of water requiring treatment.

The pump and recharge concept was used at SRS between 1998 and 2003 and at Brookhaven National Laboratory between 1997 and 2000 and again in 2007. The 1999 evaluation report (DOE/RL-99-42) discussed in detail the methods used at SRS and Brookhaven.

The pump and recharge at SRS was stopped in mid-2003 because of the high cost of groundwater treatment ($0.012 per L or $0.047 per gal). The pump and recharge resulted in a 50 to 70 percent reduction of the tritium concentration (initially up to 50,000,000 ρCi/L or 10 times the maximum tritium concentration in groundwater at Hanford) in the plume after 5 years of pumping and upgradient recharging. Operating cost for the pump and recharge was about $50,000/day. Pump and recharge at Brookhaven was stopped in 2000 because tritium levels in the vicinity of the extraction wells decreased to below the average minimum detection limit of the Brookhaven analytical services laboratory (343 ρCi/L).

The decrease was a result of the combined effects of radioactive decay, dilution, and dispersion. The Brookhaven pump and recharge system was placed in standby in 2000 and was reactivated in 2007. In addition to the pump and recharge scheme, low flow extraction of highly contaminated groundwater was undertaken at Brookhaven during 2000 and 2001 to accelerate cleanup of the plume. In total, 95,000 gal of groundwater were pumped from 10 temporary wells. The water was transported offsite for disposal (DOE/RL-2009-18).

3.3.1.1 Pump and Recharge – Application at the Hanford Site

As stated in the 1999 report (DOE/RL-99-42), groundwater pumping at the 20,000 ρCi/L concentration front would cover a distance of over 25 miles. The large distance of the front, the number of wells which would be required, and the large volume of water which would have to be pumped and potentially treated to meet applicable state and federal limits except for tritium preclude this concept from being economically feasible.

An additional factor that makes the concept not feasible at the Hanford Site is the increase in volume that would need to be pumped and potentially treated each year. Although the recharge rate is low, the additional amount of water to pump and potentially treat each year would increase significantly because of the large volume of the contaminated plume.

3.3.2 Barrier Formation

Two types of subsurface barriers have been demonstrated at DOE sites. The frozen soil barrier concept was discussed in detail in the 1999 evaluation report (DOE/RL-99-42).

Another barrier technology, Viscous Liquid Barrier (VLB), was selected by the Brookhaven National Laboratory for groundwater remediation near the Brookhaven Linear Accelerator Isotope Producer (BLIP). VLB technology was developed at Lawrence Berkley National Laboratory with DOE funding (EM-50). It used low-pressure permeation grouting to deliver a colloidal-silica grout to the subsurface. The grout gels in place and forms a barrier to liquid movement. VLB, in conjunction with a gunite cap around the surface soil of the BLIP and storm water controls, would minimize the volume of surface water percolating through the contaminated soils to the groundwater. The estimated volume of soil to be treated is approximately 110 yd^3. Modeling results showed that if the flow velocity though the activated soil is reduced to less than 1 cm/yr, short-lived isotopes including tritium will not reach the aquifer at levels exceeding the DWS (DOE/RL-2009-18).

The VLB installation was completed in 2000 at a cost of about $436,000. The cost included site characterization, grout compatibility and optimization testing, modeling, and barrier integrity verification and planning documents. According to groundwater monitoring data, the actions taken to date have been highly effective in eliminating the BLIP source of groundwater contamination. Evaluation of barrier performance is continuing. DOE, the U.S. Environmental Protection Agency, and New York State have recently agreed that "continued inspections, and maintenance of the cap, groundwater monitoring, and institutional controls in addition to the previously completed work (summarized above) are sufficient to support the selection of no further action" (DOE/RL-2009-18).

At SRS, a sheet pile dam has been used as a barrier since 2000. This barrier was designed to reduce tritiated water discharges to Fourmile Branch and has successfully reduced tritium concentrations in water discharged to Fourmile Branch by 65 percent (DOE/RL-2009-18). This dam resulted in tritiated water accumulation by a retention pond. This water is treated by phytoremediation (Section 3.3.4.1).

3.3.2.1 *Barrier Formation - Application at the Hanford Site*

Application of soil barrier technology is not economically feasible for tritium remediation in bulk groundwater because of the large volume of subsoil that would have to be frozen or injected with grout. Use of sheet pile barriers is also not applicable to the Hanford situation.

3.3.3 Decay in Slow Moving Groundwater

The 12.3 year half-life of tritium allows storage to be considered as a mitigation method. Reduction of tritium content versus storage time is illustrated in Table 3.

Table 3. Tritium Decay Versus Storage Time

Storage Time	Tritium Reduction (Percent)	Decontamination Factor (DF)
41 years	90	10
81 years	99	100
122 years	99.9	1,000

One approach for mitigation of tritium in wastewater is to store it underground in a stagnant or slow moving aquifer where it will be isolated from the general environment and human consumption for an extended period of time. This approach has the advantages of low cost and low energy consumption, provided a suitable storage location is available.

3.3.3.1 Decay in Slow Moving Groundwater – Application at the Hanford Site

Since 1995, the SALDS has received tritium contaminated effluents from the ETF. Tritium in this effluent is mitigated by storage in slow moving groundwater to allow extended time for decay before the water reaches the site boundary. This method isolates the tritium from the general environment and human contact until it has decayed to acceptable levels.

The SALDS is located just north of the 200 West Area on the Hanford Site and began receiving tritiated wastewater from the ETF in December 1995. Treated wastewater from the ETF meets all applicable state and federal limits except for tritium (Ecology, 2000). The majority of the tritium comes from processing liquid mixed wastes from single-shell and double-shell underground storage tanks and other radioactive miscellaneous wastes from the Hanford Site. The permitted average monthly flow rate is 0.25 million gal/day (90 million gal/year).

The ST-4500 permit condition S.10 requires a tritium tracking and groundwater monitoring plan. DOE has agreed to monitor the tritium plume created by the ETF discharge and update models used to predict travel time to the Columbia River (SWG-53569).

The ST-4500 permit fact sheet indicates that computer model results predict that tritium bearing effluents discharged to the ground at SALDS will take an extended period of time to travel with groundwater beneath the Hanford Site before ultimate discharge to the Columbia River. Models and discharge scenarios indicate that tritium above the DWS will not reach the Columbia River in detectable quantities (Ecology, 2000). During the long residence time in the aquifer, most of the tritium will decay to nonradioactive helium.

Discharges from the ETF to the SALDS are listed in Table 4 by date, volume, and concentration of tritium. The annual average tritium concentration may be misleading because tritiated water comes from

sources with widely different concentrations and volumes. Typically, groundwater treated at the ETF contains <50,000 ρCi/L, while K-Basin and 242-A Evaporator wastewater treated at the ETF has much higher tritium contamination (<600,000 ρCi/L since 2009). Table 5 lists tritiated water discharged to the SALDS by source for the past 5 years.

Table 4. Tritiated Water Discharges to the State-Approved Land Disposal Site

Calendar Year	Volume		Average Tritium Concentration	Tritium
	(L)	(gal)	(ρCi/L)	(Ci)
1995	2,237,000	591,000	6,200,000	14
1996	28,630,000	7,564,000	7,500,000	215
1997	57,769,000	15,263,000	610,000	35
1998	107,199,000	28,322,000	290,000	32
1999	87,313,000	23,068,000	100,000	9
2000	91,315,000	24,125,000	230,000	21
2001	98,117,000	25,923,000	630	0.1
2002	83,540,000	22,071,000	102,000	9
2003	95,655,000	25,880,000	51,000	5
2004	106,976,000	28,254,000	134,000*	15
2005	22,986,000	6,071,000	102,000*	2
2006	18,144,000	4,792,000	1,110,000*	20
2007	32,887,000	8,686,000	883,000*	29
2008	67,748,000	17,899,000	7,000*	1
2009	82,642,000	21.834,000	7,400*	1
2010	71,389,000	18,861,000	85,600*	7
2011	75,168,000	19,859,000	18,000*	1
2012	35,786,000	9,455,000	90,300*	4
2013	30,422,000	8.037,000	361,000*	11

* Weighted average

Table 5. Sources of Tritiated Water to SALDS

Calendar Year	Type*	Volume		Average Tritium Concentration	Tritium
		(L)	(gal)	(pCi/L)	(Ci)
2009	G	82,642,000	21,834,000	7,400	1
2009	K	*	0	0	0
2010	G	62,391,000	16,484,000	54,900	4
2010	K	8,998,000	2,377,000	335,300	3
2011	G	73,040,000	19,299,000	11,600	1
2011	K	2,122,000	561,000	235,000	1
2012	G	29,003,000	7,663,000	16,200	1
2012	K	6,783,000	1,792,000	450,000	3
2013	E	11,512,000	3,041,000	105,500	1
2013	K	18,910,000	4,996,000	516,000	10

* No K-Basin or 242-A Evaporator wastewater was processed through ETF during 2009.

E = ERDF leachate

G = Groundwater

K = K-Basins and 242-A Evaporator; some mixing between wastewater types can occur when switching between campaigns

3.3.4 Evaporation

One approach for disposing of wastewater and groundwater is evaporation with dispersion into the atmosphere. Potential evaporation methods include the following:

- Boiling or mechanical evaporation. A variety of industrial equipment is available for evaporation by boiling, including conventional indirect contact types (e.g., boilers and evaporators) and direct contact types (e.g., incinerators).
- Solar evaporation. Solar heat and movement of air are used to evaporate water.
- Contact with an air stream. For example, air can be bubbled through the water or water can be sprayed into the air.
- Phytoremediation. Plants take in the contaminated water through their roots and the majority of the water (including tritium) is evaporated. A fraction of the tritium is also retained in the plant matter until it decays. If the water source is at or near the surface, the plant may remove water directly from the source. Alternatively, the water may be pumped to the plants (i.e., use of contaminated water for irrigation).

Three evaporation methods (solar evaporation, irrigation, and mechanical evaporation) were considered in the 1994 Hanford Site evaluation (DOE/RL-94-77). Of 11 tritium mitigation methods evaluated, these three received the lowest rankings (Table 7-1 of DOE/RL-94-77). As discussed in the following subsection, incineration and phytoremediation have recently been selected at other DOE sites for disposal or mitigation of wastewater with trace tritium contamination.

3.3.4.1 Phytoremediation

Phytoremediation is the process of using plants to remediate contaminated areas, including soil, groundwater, and surface water (PHYTO-3, *Phytotechnology Technical and Regulatory Guidance and Decision Trees, Revised*). The process is being used at the Argonne National Laboratory (ANL) and SRS for tritiated water.

In phytoremediation, the plants take up nutrients from the contaminated soil and use the contaminated water to grow. The plants can breakdown, trap, and hold contaminants in the leaves and/or stems and transpire the water into the atmosphere as part of natural plant growth. The transport of water in soil and vegetation and accumulation of biomass in forest trees are considered the most important physical, chemical, and biological transport processes for estimating partitioning of isotopes to vegetation and the amount of fixed tritium in a forest that has been exposed to tritiated irrigation water (DOE/RL-2009-18).

The turnover time of tritiated water in the conducting outer rings of the roots and stem and in the leaves is on the order of hours to several days. The turnover time of tritiated water in the inner, older rings of the roots and stem is on the order of days to years depending on the size of the tree. Approximately 60 percent of the tritium fixed during synthesis remains in the biomass until released by decay or combustion after death of the tree. The remaining 40 percent will be exchanged with hydrogen in water (DOE/RL-2009-18).

As mentioned, phytoremediation at the ANL is being performed on a site that is contaminated with tritium. The goal of the installation was to protect downgradient surface water and groundwater by intercepting the contaminated groundwater with tree roots, removing moisture from the upgradient soil area, reducing water infiltration, preventing soil erosion, degrading and/or transpiring the residual volatile organic compounds, and removing tritium from the subsoil and groundwater (ANL/ES/RP-66172, *317/319 Phytoremediation Site Monitoring Report - 2009 Growing Season*). The phytoremediation system was designed so that in a few years, the rate of transpiration of water will match the natural recharge to the aquifer. This will essentially stop the movement of the contamination plume. Work on phytoremediation has continued at the ANL, with an anticipated operating period of 20 years (DOE/RL-2009-18). Based on recent sampling results, remedial actions implemented in the 1990s appear to have been effective at preventing any further discharge of tritium (ANL-13/02, *Site Environmental Report for Calendar Year 2012*).

The purpose of the tritium phytoremediation project at SRS is to reduce the volume of water reaching contaminated aquifers and reduce the volume of contaminated water reaching surface water sources. Reduction of recharge and removal of water via the plant roots increases the time for decay of the tritium while it is still in the ground. Although this results in tritiated water being transpired into the atmosphere, this remediation method is considered acceptable at SRS because of the distance to the nearest population center (DOE/RL-2009-18). Dose studies associated with this treatment system indicate that the exposure to site workers and offsite maximally exposed individuals is very low, approximately 6 mrem/year and 0.004 mrem/year, respectively (SRNS-MS-2012-00198, *Forest Irrigation of Tritiated Water: A proven tritiated water management tool*).

A sheet pile dam for collection of water and forest irrigation system was installed at SRS in 2000 and 2001, respectively, for tritium remediation at a cost of about $1,500,000. Tritiated water behind the dam is used for irrigation of 25 acres of natural forest pines and hardwood trees located upgradient of the seep line. The irrigation supplements natural precipitation. Annual operating cost is about $500,000 and about 2,000,000 of tritiated water was used for irrigation of the forest in FY 2003. This project has resulted in a 65 percent reduction in tritium concentrations in water discharged to Fourmile Branch (DOE/RL-2009-18).

3.3.4.2 Evaporation – Application at the Hanford Site

Evaporation has the advantage that it may completely eliminate the liquid tritium waste stream, often at a moderate cost compared to separations options. A major negative is that it discharges the tritium into the air. Also, phytoremediation for minimizing water entering a contaminated aquifer typically uses either hybrid poplars or pine trees. Neither of these types of plants would be amenable to the natural environment on the Hanford Site. The 1994 tritium technology evaluation (DOE/RL-94-77) scored irrigation (phytoremediation) as the poorest based on relative cost and risk. Table 6 shows the maximum tritium concentrations at various locations on the Hanford Site.

Table 6. Maximum Tritium Concentrations at Hanford

Area or Operable Unit	Maximum Tritium Concentration (ρCi/L)
100-BC-5	21,000
100-HR-3-D	19,000
100-HR-3-H	5,900
100-FR-3	2,500
100-KR-4	130,000
100-NR-2	35,000
200 ZP-1	200,000
200 UP-1	350,000
200 BP-5	35,000
200 PO-1	610,000
300-FF-5	1,100,000
1100-EM-1, North Richland	151

Source: DOE/RL-2013-22, *Hanford Site Groundwater Monitoring Report for 2012.*

3.4 Relative Cost of Mitigation Options

Costs for options to mitigate tritium in Hanford wastewater will vary substantially, depending on the technical process option selected. Costs can also be expected to vary substantially depending on site specific and project specific factors. Available cost estimate information for various technologies is identified in the sections that discuss those technologies. In most cases, the available cost data were not developed for specific Hanford wastewater treatment scenarios. However, the available data provides some insight into the rough order of magnitude costs.

To develop more accurate cost estimates, engineering studies would need to be performed to estimate equipment size, energy demand, and capital and operating costs. Specific scenarios would need to be defined, including wastes to be treated or otherwise mitigated, location, capacity, operating duration, and applicable state and federal requirements for treated wastewater and concentrated product. Despite these limitations, some general comments on relative costs can be made, discussed as follows and summarized in Table 7:

- Separation is typically the most expensive overall mitigation option. For a large base-load type facility operated almost continuously, sized to treat the full stream currently discharged from the ETF to the SALDS, and designed to reduce tritium content below the DWS, the total treatment cost (capital, operation, utilities, and other project costs) is expected to be in the range of dollars per gallon, likely several dollars per gallon. The water distillation, CECE, and bithermal hydrogen/water processes all appear to be technically viable candidate separation processes. Available information is not sufficient to determine a clear preference or ranking among these tritium separation processes. The preferred option may vary depending on power and steam costs, plant capacity, and base load versus cyclic or campaign type operations and other scenario/site specific factors.
- Energy costs for treating high volume, low concentration tritium wastewater by conventional separation technologies (e.g., distillation, electrolysis, and CECE) have been estimated to range from 50 to 200 MW (DOE/RL-2009-18). The energy component to the treatment cost would range from $0.50 to $2.00/gallon, and the total power requirement would consume 5 to 20 percent of the electrical capacity of the Energy Northwest Columbia Generating Station. The high energy consumption is not deemed appropriate for removing a small amount of tritium from the Hanford ETF outfall.
- For separations options that produce a concentrated tritium product, options for handling, storage, and disposal will also significantly affect total cost.
- Cost estimates prepared to support selection of the current SALDS option suggest that the cost for mechanical evaporation will be on the order of tenths of a dollar per gallon, while costs for ground discharge and extended delay in slow moving groundwater (current SALDS approach) are estimated to be less than $0.01/gal. These estimates were based on the assumption that the full stream currently discharged to SALDS would be treated.
- The cost for all mitigation options will tend to increase in terms of dollars per gallon for smaller processing capacity and intermittent operation.
- Costs for water treatment at existing Hanford Site pump and treat projects provide additional perspective on typical wastewater treatment costs. Treatment cost for these projects is less than $0.50/gal.

- For wastewater that is already in the ground, decay in place appears to be the obvious lowest cost option for tritium mitigation. For wastewater discharged from the ETF, continued discharge to groundwater via the SALDS is undoubtedly the lowest cost mitigation option. The tritium concentration in groundwater will gradually drop due to radioactive decay and dilution. For the contamination levels in SALDS discharge and in groundwater identified at Hanford, decay alone will reduce tritium concentrations below the DWS in less than 100 years.
- Costs for options such as pumping, underground barriers, and phytoremediation may be relatively low in terms of dollars per gallon but are highly project specific. These options do not appear to be applicable to the Hanford wastewater treatment application.

For mitigation of relatively small volumes, such as treatment or relocation of small volume, high concentration wastes, mitigation process costs (equipment and operation) are likely to be overshadowed by other project costs such as waste characterization, engineering, technology development/definition, safety evaluations and approvals, permitting, and overhead.

Table 7. Comparison of Mitigation Approaches

Mitigation Approach	Typical Implementation or Technology	Efficiency	Cost Information	Comments
Separation treatment	Water Distillation, CECE, bithermal hydrogen	Efficiencies to meet drinking water standard demonstrated for CECE, expected achievable by others with sufficient staging	For large base load type facility costs expected to be in dollars per gallon range; increased by smaller size, intermittent operation, or short operating life	Handling and disposal of concentrated tritium stream will result in additional costs
Decay	Discharge to slow moving ground water via crib or percolation pond	Efficiency determined by travel time to release point; models predict acceptable efficiency for SALDS	For large volume operated over a long period of time (such as SALDS) cost expected to be in cents per gallon range	Current SALDS approach
Decay	Upgradient pumping, barriers, or reduced inflow to delay release	Site specific and highly variable	Costs are site specific and highly variable. Massive barriers to control ground water movement at Hanford judged economically infeasible	Does not appear applicable to Hanford due to large areas and large perimeter
Mechanical evaporation	Boiler	100% of tritiated water can be evaporated	For large volume base load type, facility cost is expected to be in tenths of a dollar per gallon range; expected to be much higher for small volumes	Results in atmospheric discharge; rated very poorly in initial option evaluations
"Natural" evaporation	Solar evaporation, evaporation by plants, irrigation	Solar evaporation could evaporate essentially 100%, others highly variable	Costs are site specific and highly variable	Results in atmospheric discharge; rated very poorly in initial option evaluations

CECE = combined electrolysis catalytic exchange

SALDS = State-Approved Land Disposal Site

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