Fukushima Light Water Detritiation System

Water Distillation Option


Presented at the 34th Tritium Focus Group Meeting on September 23-25, 2014, Idaho National Laboratory, Idaho Falls, Idaho
400 m$^3$/day is accumulation rate. Assume 500 m$^3$/day capacity is built to also process backlogged water over a number of years.
# Water Distillation (WD) vs Combined Electrolysis & Catalytic Exchange (CECE) – Energy Cost Big Picture

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Units</th>
<th>WD</th>
<th>CECE</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>H/T Separation Factor for Light Water Detritiation</td>
<td>-</td>
<td>1.059 @ 50°C</td>
<td>5.017 @ 60°C</td>
<td>CECE separation factor is not the only important consideration.</td>
</tr>
<tr>
<td>Boilup or Electrolysis Energy for Reflux</td>
<td>kWh per kg of H₂O</td>
<td>0.66</td>
<td>5 to 6</td>
<td>Energy of electrolysis is much higher than energy of boilup.</td>
</tr>
<tr>
<td>COP* for Energy Utilization</td>
<td>Energy multiplier by heat pump or fuel cell</td>
<td>8 to 10 With Heat Pump</td>
<td>1.3 With Fuel Cell</td>
<td>WD recycles energy much more efficiently.</td>
</tr>
<tr>
<td>Energy demand for 500 m³/day throughput</td>
<td>MW(e)</td>
<td>25 to 40</td>
<td>155 to 210</td>
<td>WD uses much less energy.</td>
</tr>
</tbody>
</table>

*CECE large Separation Factor does not mean low energy consumption!

Even if CECE separation factor was infinity, CECE would require 104 to 125 MW(e) power for 500 m³/day throughput – about 4 to 5 times more than WD.

*COP = coefficient of performance. For CECE the practicality of energy recovery/recycling is questionable.
## WD vs CECE – Hazards Evaluation

<table>
<thead>
<tr>
<th>Parameter</th>
<th>WD</th>
<th>CECE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrogen Explosion Hazard</td>
<td>No.</td>
<td>Yes</td>
</tr>
<tr>
<td>Pure Oxygen Hazard</td>
<td>No.</td>
<td>Yes</td>
</tr>
<tr>
<td>Leakage Potential (during normal operation)</td>
<td>Vacuum process – leaks don’t result in emission</td>
<td>Above atmospheric process. Greater potential for tritiated hydrogen/water vapor leak</td>
</tr>
<tr>
<td>Tritium Inventory</td>
<td>Small. Falling film reboiler has small inventory (less than 0.5 hour residence time). Most of system inventory is in the column packing at much lower tritium concentration.</td>
<td>Large. Electrolyser has large liquid inventory (about 16 hours residence time), much larger than WD reboiler. Electrolyser contains tritium at highest concentration.</td>
</tr>
<tr>
<td>Electrolyte handling</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Operational complexity and potential for operator error</td>
<td>Simple process</td>
<td>Relatively complex process</td>
</tr>
</tbody>
</table>

*Both WD and CECE can be operated safely. However, WD has better Inherent Safety.*
Note: External tritiated water accepted by OPG for disposal is sent to the station HW upgrader to recover D2O and DTO. H2O is separated and sent to drain.

Darlington TRF (4 CD Columns)

Station Heavy Water Upgrader (3 WD Columns in series)
WD Industrial Experience: $^{18}\text{O}$

- WD is used to produce $^{18}\text{O}$. There are several producers in the world.

- CIL in Xenia, Ohio has been producing $^{18}\text{O}$ water for the PET community since 1996. The expanded plant produces 250 kg/yr of $^{18}\text{O}$ used for production of $^{18}\text{F}$ in the PET industry, and for other uses in medical research.

- Fukushima WD columns could produce $^{18}\text{O}$ in addition to tritium recovery, to offset capital cost and to produce a medically valuable isotope that benefits the medical community.

- Cambridge Isotope Laboratories in Xenia, Ohio produces $^{18}\text{O}$ by water distillation using surplus Heavy Water Upgrader columns. Water distillation is the most common technology used for separating Oxygen isotopes.
GEH-C Water Distillation Experience 1

- GEH-C has developed a competitive CY type WD packing.
- Packing surface activation is key to separation performance.
- Surface wetting requires a cupric oxide film.
GEH-C Water Distillation Experience 2

- Design of several WD Light Water Detritiation Facilities

- This example shows *Mechanical Vapor Compression* for ~90% heat recovery

- 500 kg/h throughput with ~1 MW(e) power
Heavy water upgraders in Canada use dynamic simulation to analyze both deuterium and tritium separation in water distillation.

- Note that the tritium activity in the head product is very low.

- Heavy water upgraders also detritiate water!

Reference: Ram Davloor, Gretel Steinberg, Anthony Busigin, Salwan Saeed, “Major Rehabilitation of the Bruce A Heat Transport Heavy Water Upgrader and Effect on Overall Upgrader System and Tower Packing Performance”, 9th CNS International Conference on CANDU® Maintenance, Toronto, Ontario, Canada, December 4-6, 2011
WD Energy – Simple Heating & Cooling

Energy Requirement is 13.68 kW per kg/h of feed.

Feed rate of 500 m³/day corresponds to 285 MW(t)

- Simple heating and cooling has high energy demand due to high reflux ratio for H₂O/HTO separation – but less energy demand than with H₂O/HDO separation.
- Canadian heavy water upgrader WD columns use inexpensive low pressure turbine exhaust steam.
- Fukushima could supply natural gas reboilers for simple heating – but heat pumping to reduce energy demand can significantly reduce energy cost.
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WD Energy – Heat Pump with Hot-Cold Water Circuits – Option 1

- Industrial Heat Pump, ammonia refrigerant, pumps heat from 38°C to 62°C
- Standard Heat Pump packages available

Energy Requirement is 1.7 kW per kg/h of feed

About 87% of heat energy is recycled within the system

Feed rate of 500 m³/day corresponds to 35 MW(e)
About 90% of system cost is in the large Stripping Section equipment. The rest of the equipment is small.

**Stripping Section**
(about 18m packing height)

**Tritiated Light Water**
400 m$^3$/day
- $x[H] = 0.99985$
- $x[D] = 0.00015$
- $x[T] = 8.39E-12 (2.7E-05 Ci/kg)$
- $x[18O] = 0.99758$
- $x[17O] = 3.77E-04$
- $x[16O] = 2.04E-03$

Steady state 400 m$^3$/day case is shown. The same system can detritiate water at 500 m$^3$/day at about 30% lower $^{18}$O production rate.

**Enriching Section**
(about 53m packing height. Only 18m is necessary for tritium separation. Extra packing height for $^{18}$O separation generates significant income.)

**WD Tapered H/T Separation Column**
(DF = 104)
Volume Reduction Factor = 6000
Intermediate Reboiler – boils up 98% of liquid

Extra length for $^{18}$O separation is shown in green. Due to small diameter, the incremental cost for separating $^{18}$O is minimal.

**WD Volume reduction of 6000 produces 85% D$_2$O, with 0.14 Ci/kg, 67 kg/day**

- **$^{18}$O is easily separated from tritiated water and upgraded to 99%**.
  Nominal value of H$_2^{18}$O is $100/gram$.

- **Small volume of tritiated D$_2$O can be shipped to a Canadian heavy water reactor station for use as D$_2$O make-up. (Heavy water reactors require 2 to 4 tonne/year of D$_2$O make-up to compensate for losses.)**

**$^{18}$O Production is 16 kg/day at 400 m$^3$/day throughput or 11 kg/day at 500 m$^3$/day throughput. ($^{18}$O production increases at lower detritiation rate due to higher reflux ratio.)**

**Value of $^{18}$O produced is > $1 million per day!**
Fukushima WD Concentration Profile

- Cascade is designed to be mostly light water.
- Heavy oxygen isotopes and deuterium rise sharply at the bottom.
- Bottom product flow corresponds to a volume reduction factor of 6000.

Significant enrichment of $^{18}\text{O}$ to 24%.

Most of system capital and energy cost (98%) is in short section of large diameter column before taper.

Shorter length of column needed for just tritium separation doesn’t enrich $^{18}\text{O}$ very much. Cost of extending the small diameter column is minimal, with no additional energy cost.

Significant enrichment of D to 87%, and T by factor of about 6000 to 0.14 Ci/kg.

Small column diameter, $1/50^{th}$ flows.
Fukushima WD System Layout 1

Water Distillation Area, Maximum Equipment Height 38m

Heat Pump Area
Maximum Equipment Height 10m

maintenance and Administrative Area
Maximum Height 10m

Product Handling Area
Maximum Equipment Height 10m

All dimensions are in units of meters.

Oxygen-18 Polishing
Columns and Pumps

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Fukushima WD System Layout 2

Approximate Water Detritiation Building Profile
Conclusions

✓ WD technology is proven, including scale-up to large diameter
✓ WD is a simple, safe and reliable process
✓ WD design & simulation codes are proven:
  • steady state
  • dynamic
✓ Industrial Heat Pumps are available for recycling heat
✓ $^{18}O$ byproduct will offset or possibly pay for cost of water detritiation
✓ $^{18}O$ production benefits medical diagnostics

We believe WD is the most proven and economic solution for Fukushima water detritiation

Thank you for your attention!
Appendix
Presentation Outline

1. Fukushima Light Water Detritiation Requirement
2. Water Distillation vs CECE – The Big Picture
3. Water Distillation Industrial Experience
4. Water Distillation $^{18}$O Byproduct Opportunity
5. GEH-C & Canadian Water Distillation Experience
6. Water Distillation Process Configuration and Energy Demand:
   a) Mechanical Vapor Compression
   b) Industrial Heat Pump
7. Conclusions
### Industrial Scale of Water Distillation

**Built 1943-1944**

- WD technology is now much more compact than in WW II era.

- Canadian heavy water reactor Heat Transport Upgraders routinely recover tritium and deuterium from mostly light water (typical feed is 70% light water, 30% heavy water, and about 1 Ci/kg tritium in the heavy water component).

- Canadian heavy water reactor Heat Transport Upgraders have been in continuous use since the 1960s, and are the most proven and successful method of large scale light water detritiation, although heavy water upgrading is their primary function.

**Savannah River individual columns were similar in size to current large CANDU® heavy water upgraders.**

**However, CANDU® upgraders use only two or three columns.**

**Morgantown, W. VA., Distillation Plant for Heavy Water**

- Savannah River WD columns Built Early 1950s.

- Used to upgrade heavy water from 20% to 90%.
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WD High Efficiency Packing – Effect on Column Height

- WW II era used inefficient bubble cap tray columns
- Structured wire gauze packing allows modern WD to be drastically smaller than historical bubble cap tray columns
- Structured packing, like that used in WD columns is used in many other industrial distillation applications at large size.

Relative Height of Distillation Columns Depending on Packing Type for Similar Overall Isotope Separation Factor

- Morgantown Type Bubble Plate Column (40 x taller than Dixon Ring Lab Column)
- BX Structured Packing (CANDU)
- CY Structured Packing (CANDU)
- Dixon Rings or Similar Packing for Lab Scale Columns
Heavy Water Upgrader Experience

- Dynamic simulation closely matches measured performance
- Dynamic simulation is used to analyze column behavior with varying feed composition, and to simulate column detritiation prior to maintenance shutdown

Reference: Ram Davloor, Gretel Steinberg, Anthony Busigin, Salwan Saeed, “Major Rehabilitation of the Bruce A Heat Transport Heavy Water Upgrader and Effect on Overall Upgrader System and Tower Packing Performance”, 9th CNS International Conference on CANDU® Maintenance, Toronto, Ontario, Canada, December 4-6, 2011
GEH-C Water Distillation Experience

- GE Healthcare built a water distillation system in the UK to recover tritium from water after thermal oxidation of tritiated organic wastes.

- Inactive commissioning demonstrated WD separation performance by separating oxygen isotopes.

Fit between dynamic simulation model and experiment is excellent. Water distillation is well understood!
Heat Pump COP Definition

COP = Coefficient of Performance

\[
COP = \frac{Q}{W}
\]

\(Q\) = Heat supplied to the process
\(W\) = Work consumed by heat pump

\[
COP_{heating} = \frac{Q_H}{W} = \frac{Q_C + W}{W}
\]

\[
COP_{cooling} = \frac{Q_C}{W}
\]

\(Q_C\) = heat removed from cold reservoir
\(Q_H\) = heat supplied to hot reservoir

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An ammonia heat pump compressor for WD is relatively small due to operation in the 13 to 30 bar pressure range.

A corresponding mechanical vapor compressor is much larger due to the low WD condenser pressure of 0.08 bar, requiring very high volumetric flow.

An ammonia heat pump is available, but it has inferior heat transfer characteristics, and is less environmentally friendly in case of environmental emissions.

The Heat Pump working fluid system is hermetically sealed, like a household refrigerator.
WD Heat Pump Maximum Theoretical Performance

- WD Column Design Characteristics:
  \[ T_{\text{condenser}} = 315K (42^\circ C), P = 8 \text{ kPa} \]
  \[ T_{\text{reboiler}} = 320K (47^\circ C), P = 11.6 \text{ kPa (intermediate reboiler)} \]
  \[ \Delta T = 5K \text{ across condenser and reboiler heat exchangers} \]

\[
\text{COP}_{heating,max} = \frac{T_{\text{hot}}}{T_{\text{hot}} - T_{\text{cold}}} \approx \frac{(320 + 5)}{(320 + 5) - (315 - 5)} = 21.67
\]

- \( \text{COP}_{heating,max} \) is for an ideal Carnot heat pump
- In a practical design, \( \text{COP}_{heating} \) of 8 to 10 is readily achievable

1 kW(e) provides 8 to 10 kW(t) of WD boilup.
**WD – Industrial Heat Pump 2**

- Typical ammonia heat pump performance
- Heat pumps are available in a wide range of sizes
- Compressor operating life is 20+ years
- Units are hermetically sealed

**Typical Ammonia Heat Pump COP (~60% Carnot Efficiency)**

- COP = 12
- COP = 10
- COP = 8
- COP = 6
- COP = 4

Water Distillation application COP ≈ 9
WD Energy – Direct Mechanical Vapor Compression – Option 2

- Mechanical Vapor Compression (MVC) drastically reduces energy cost of distillation.
- Costs associated with steam boiler system are avoided.
- Water vapor MVC is widely used in water desalination, orange juice evaporation, sugar evaporation, etc.
- Capital cost of vapor compressors is often recovered in just one to two years of energy savings

Energy Requirement is 1.4 kW per kg/h of feed

90% of heat energy is recycled within the system

Feed rate of 500 m³/day corresponds to 28 MW(e)
Industrial Scale Mechanical Vapor Compressor

- Steam compressors have been used for over 50 years
- Exceptionally high plant availabilities of 96% – 98% are reported
- MVC specific energy consumption of 7-10 kWh/ton distillate is achieved in sea water desalination application
- WD application at 0.08 bar condenser pressure requires large vapor flow of $7.7 \times 10^6$ m$^3$/h for 500 m$^3$/day detritiation

GE Steam Compressors are available from 5,000 to 250,000 m$^3$/h. See: [www.ge-energy.com/content/multimedia/_files/downloads/Centrifugal_Compressors_SRL.pdf](http://www.ge-energy.com/content/multimedia/_files/downloads/Centrifugal_Compressors_SRL.pdf)
WD Energy – Auxiliary Heat Pump Circuit – Option 3

- Auxiliary Heat Pump, ammonia refrigerant, pumps heat from 40°C to 65°C

Energy Requirement is 1.4 kW per kg/h of feed

90% of heat energy is recycled within the system

Feed rate of 500 m³/day corresponds to 28 MW(e)
WD Energy – Multiple Effect Distillation and Thermocompressor Vapor Compression – Option 4

- Relatively complex process with about 60% energy savings as compared to direct heating and cooling.
- Not as much energy savings as with MVC or Heat Pump alternatives.
- There are no mechanical compressors in this alternative, so capital cost is lower.

Energy Requirement is 6 kW per kg/h of feed.

Feed rate of 500 m³/day corresponds to 124 MW(t)
Recovery of $^{18}$O from QT$^{18}$O, Option 1

- No flammability or explosion hazard
- Exchange rate between water and carbon dioxide is well known
- Exchange is slow at room temperature but fast enough at elevated temperature to not require catalyst
- Equipment is small and inexpensive
- CO$_2$ molecule does not carry tritium
Recovery of $^{18}$O from QT$^{18}$O, Option 2

- Hydrogen flammability and explosion hazard – but safe closed system
- No electrolyser is required
- Small LPCE columns
- LPCE Catalyst is more expensive than inert packing
- $Q_2$ molecule carries tritium between columns

Hydrogen – Water Oxygen Isotope Exchange

$H_2^{16}O$ to QT$^{18}$O

QT$^{18}$O to $H_2^{16}O$ Head Product to Drain

$99\% H_2^{18}O$ Product

Q = H, D or T

$H_2^{18}O$ to final $^{18}$O Enrichment

$H_2^{16}O$ to QT$^{18}$O

80°C LPCE

35°C LPCE

$Q^{16}O$ to Tritium Disposal

~86% $D_2^{16}O$ with 0.14 Ci/kg Tritium

Small Diameter $H_2^{18}O$ Polishing Column

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**H₂¹⁸O Product Final Enrichment**

- Small diameter column to enrich H₂¹⁸O to 99% purity
- WD is economic technology for H₂¹⁸O production irrespective of tritium separation
- H₂¹⁸O product offsets cost of detritiation.

**H₂¹⁸O Enrichment Column Composition Profile**

H₂¹⁷O accumulates in the middle of the column, and could be withdrawn at a small rate as a separate product. ¹⁷O is the only oxygen isotope with nuclear spin, making it valuable in medical Magnetic Resonance Imaging (MRI).
Water Distillation $^{18}$O Byproduct Opportunity

<table>
<thead>
<tr>
<th>Stable Oxygen Isotope</th>
<th>Natural Abundance</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{16}$O</td>
<td>99.758%</td>
</tr>
<tr>
<td>$^{17}$O</td>
<td>0.038%</td>
</tr>
<tr>
<td>$^{18}$O</td>
<td>0.204%</td>
</tr>
</tbody>
</table>

- Almost all the $^{18}$O produced in the world is produced by water distillation
- Market value of H$_2^{18}$O is about $100/gram

- $^{18}$O is the precursor for $^{18}$F used in Positron Emission Tomography (PET) medical imaging scanners
- $^{18}$F is produced by irradiation of $^{18}$O with high energy protons (18 MeV) from a cyclotron. $^{18}$O + $^1$H $\rightarrow$ $^{18}$F + n. The target material is H$_2^{18}$O.
- $^{18}$F is a positron emitter with a half life of 109.77 minutes.