Light Water Detritiation using the CECE Process

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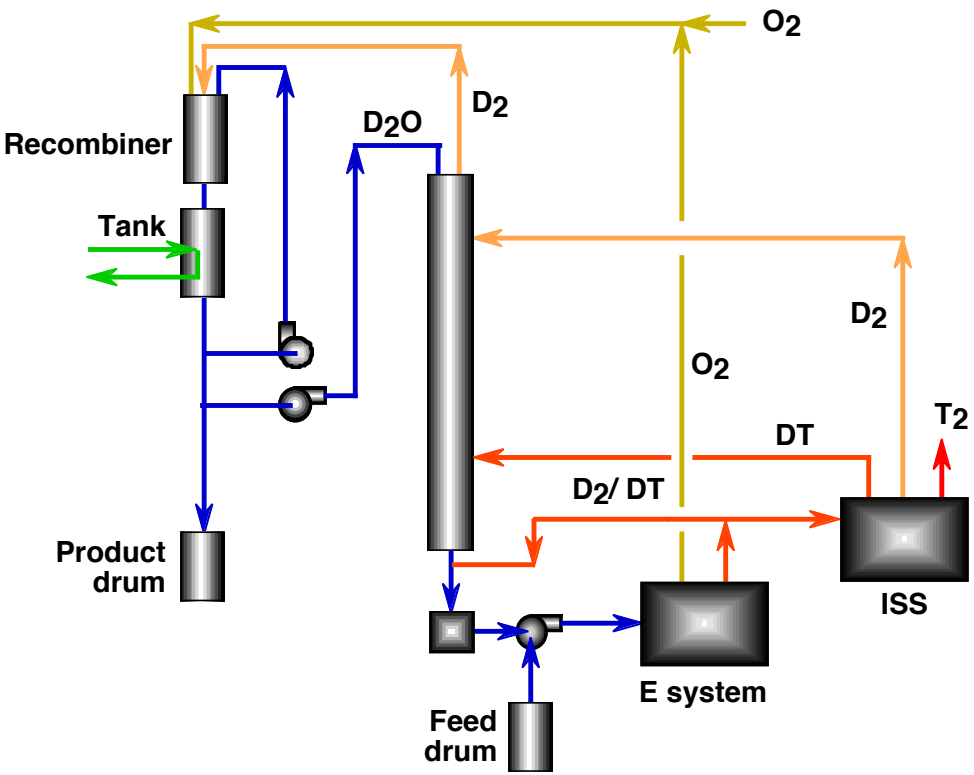
Chronology of installing a CECE Facility at NSSI

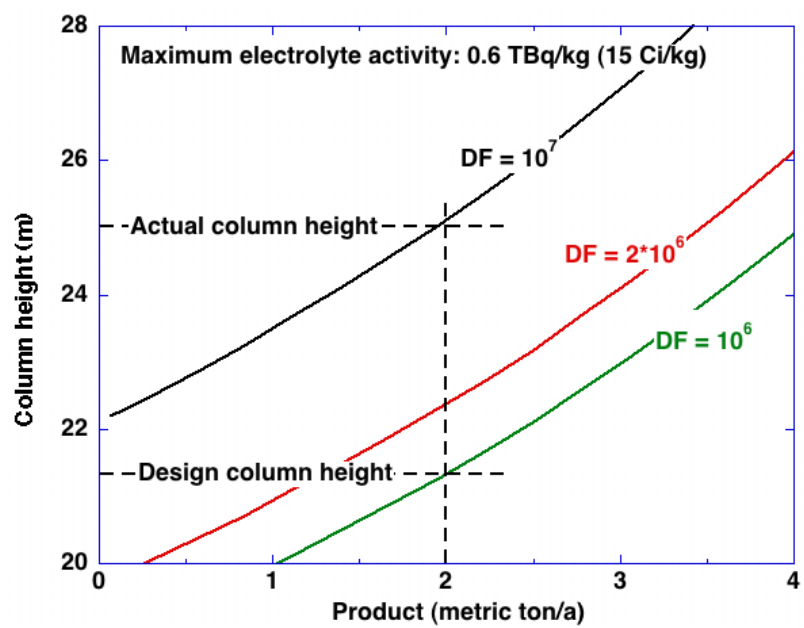
2003	CECE concept to presented at the 8th International Symposium on "The Synthesis & Applications of Isotopes & Isotopically Labeled Compounds" in Boston, Massachusetts
2004	The performance of a 5.6 m, 3.6 cm diameter column to concentrate HTO is presented at 25th Tritium Focus Group, Germantown, MD
	A conceptual design to detritiate 2 tonnes/year of heavy water is presented at 26th Tritium Focus Group, Aiken, NC and the 7th International Conference on Tritium Science and Technology, Baden-Baden, Germany
2005	Heavy water detritiation facility commissioned
2007/8	Processed 800 L of heavy water with activities ranging from 70 μCi/kg to 2 Ci/kg
2011	Reconfigured facility to process light water
2013	Performance tests using low activity light water completed
2014	System design to increase throughput completed
2015	Preliminary high throughput test initiated

The heavy water CECE detritiation facility comprises four subsystems: Electrolyzer, Isotope Separator, Liquid-phase catalytic exchange (LPCE) columns, and Recombiner

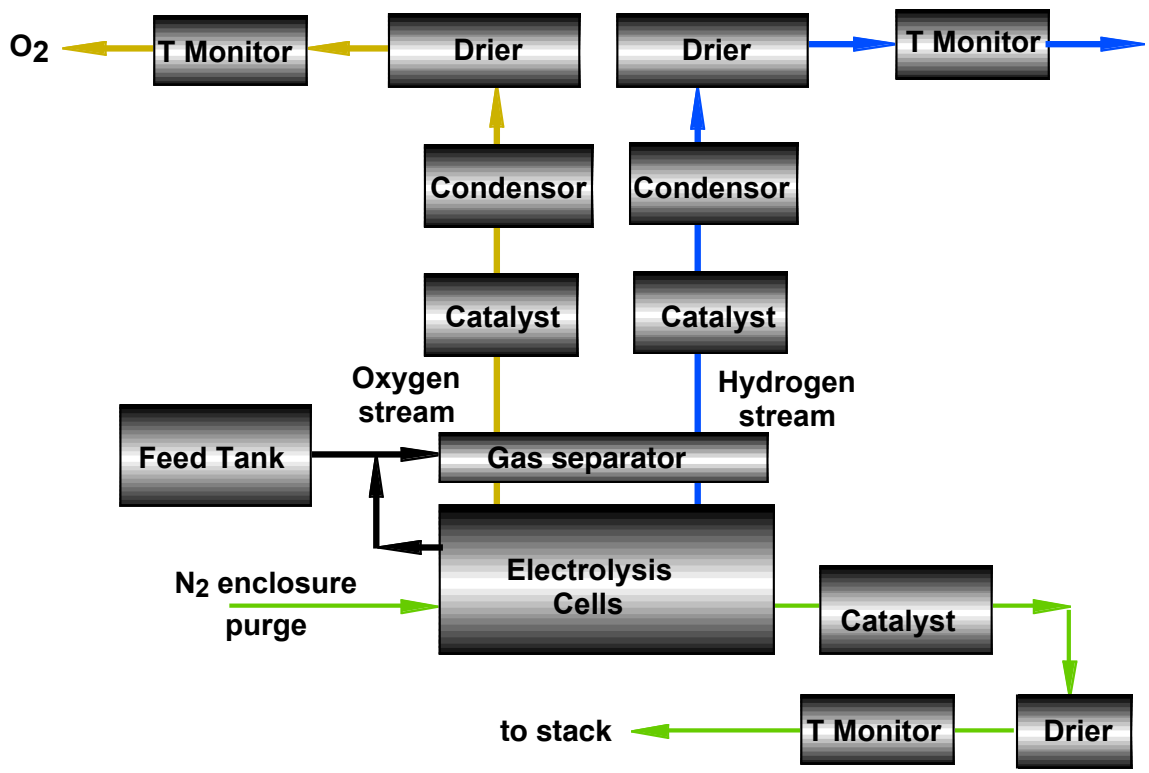


Facility design parameters

- Detritiation factor (DF) > 1 * 10^6
- Product activity < 5 μCi/kg
- G/L = 1.05
- Production rate: 1.8 tonnes/a
- Duty factor: 290 days



A commercial 5.8 liter/hour electrolyzer was modified for tritium service.



H_2 production:

- Flow Rate: 120 sLPM
- Pressure: < 140 psig
- Dew Point: < - 60 °C

O2 production:

- Dew Point: < - 80 °C

Max Electrolyte Activity:

- 15 Ci/kg
- No degradation

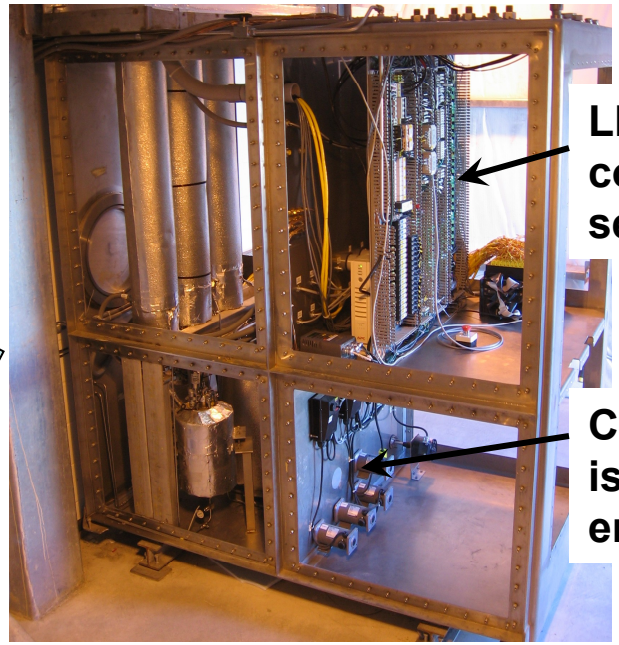
Water process rate: 30.3 tonnes/a, 90% plant efficiency, $\lambda = 3$

Columns, control systems and recombiner are housed in separate ventilated enclosures for safety

Three 8.3 meter column sets are housed in an insulated and ventilated enclosure with room for additional columns

D_2/O_2 recombiner & cooling tank are located in a separate enclosure above the roof line.

Electrolyzer behind wall in tritium lab.

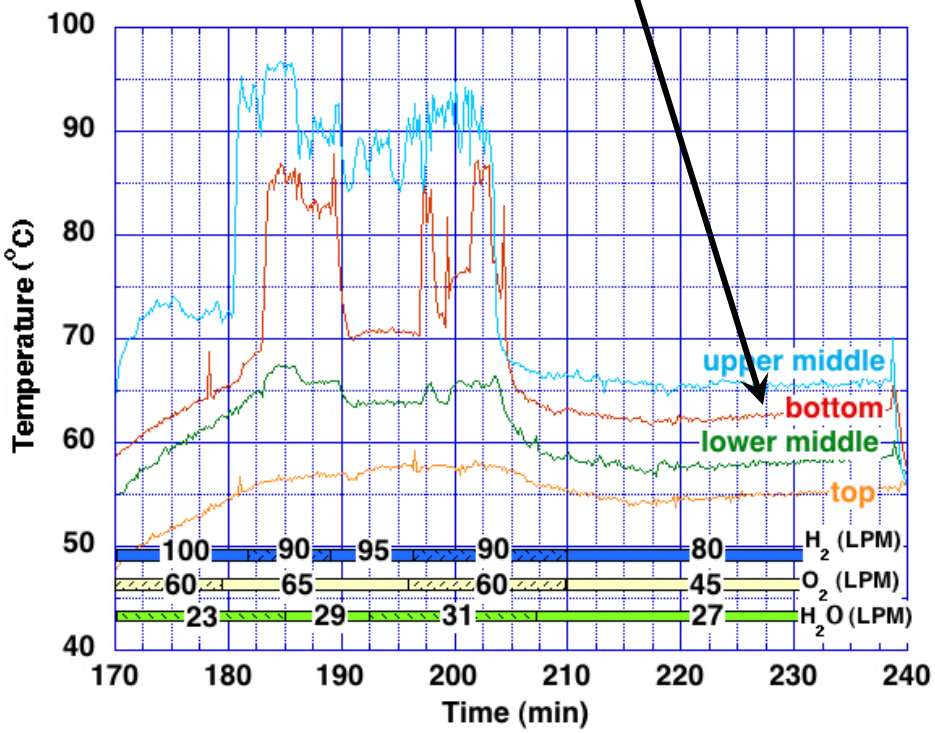


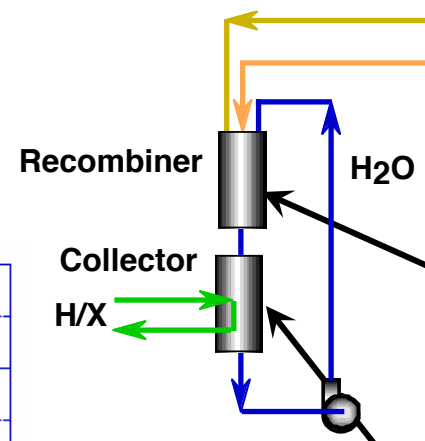
LPCE & recombiner control systems in a separate room

Control systems isolated from H_2 environment

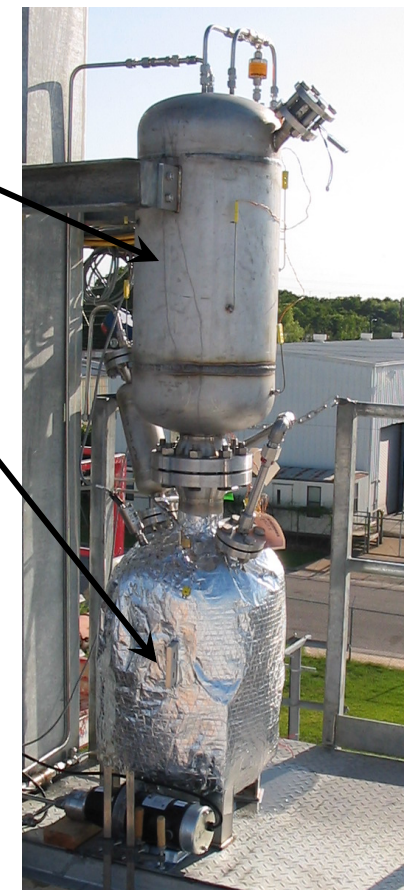
H_2 oxidation at 95 sLPM in the Recombiner is demonstrated. D_2 oxidation at 50 sLPM is routine.

16 kW of heat are removed in steady state at 80 sLPM

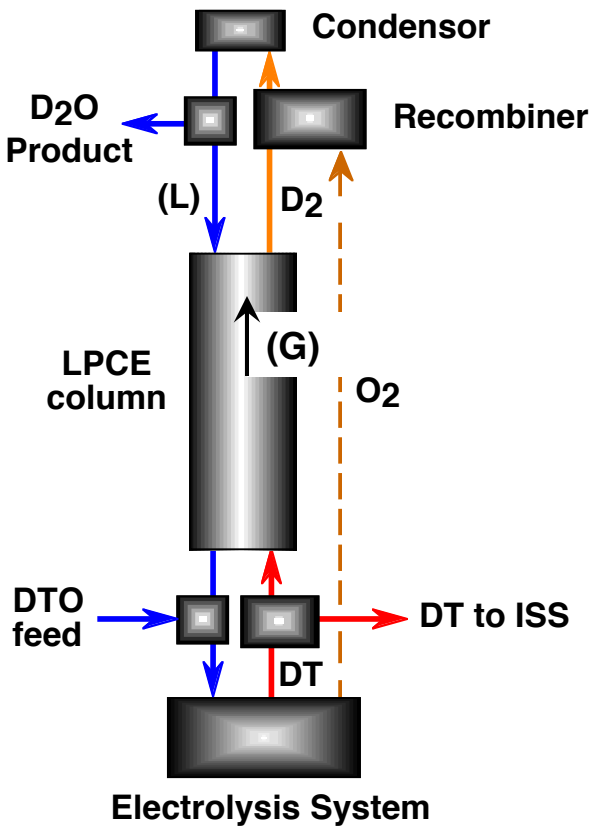


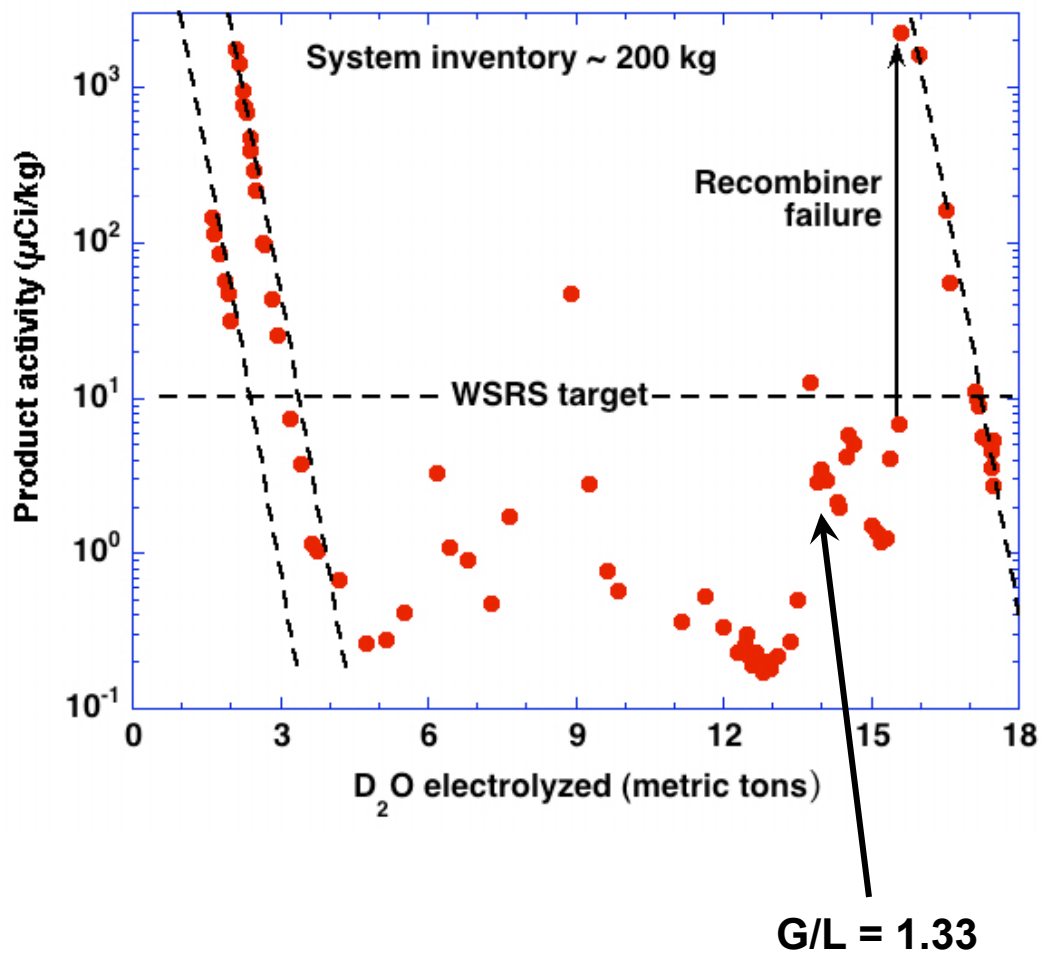


Optimum operating temperature range

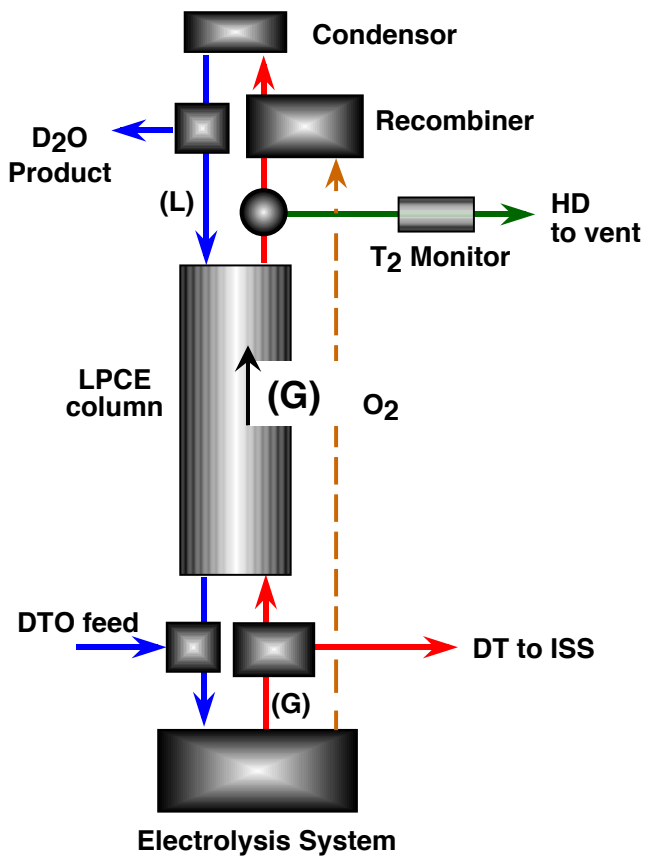


The LPCE column detritiation factor reached $3*10^7$ with a gas (G) to liquid (L) ratio of 1.05





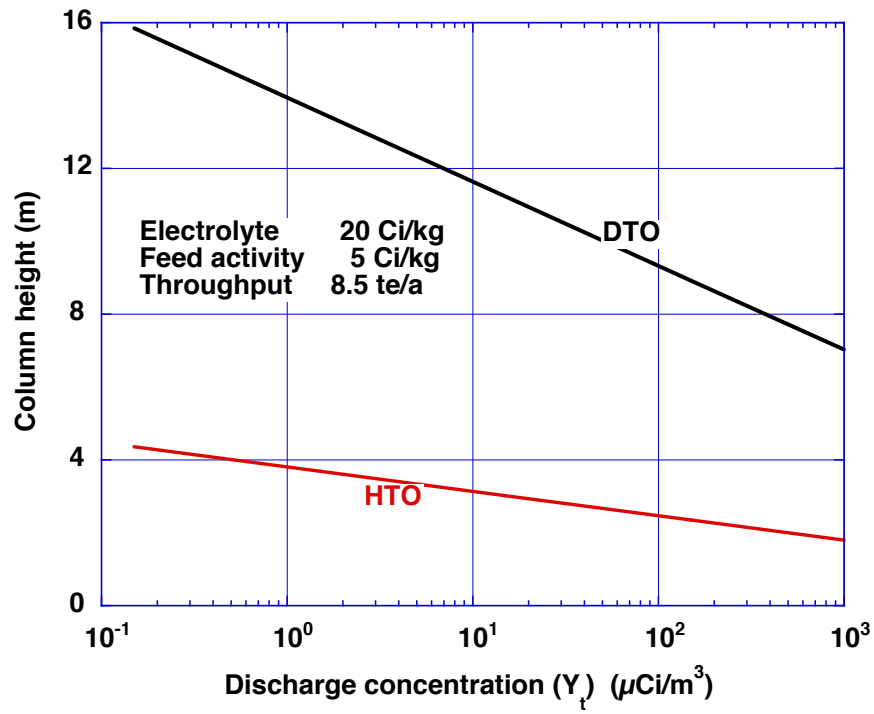
The separating power of columns is greater for HTO than DTO. The columns are shorter when processing HTO.



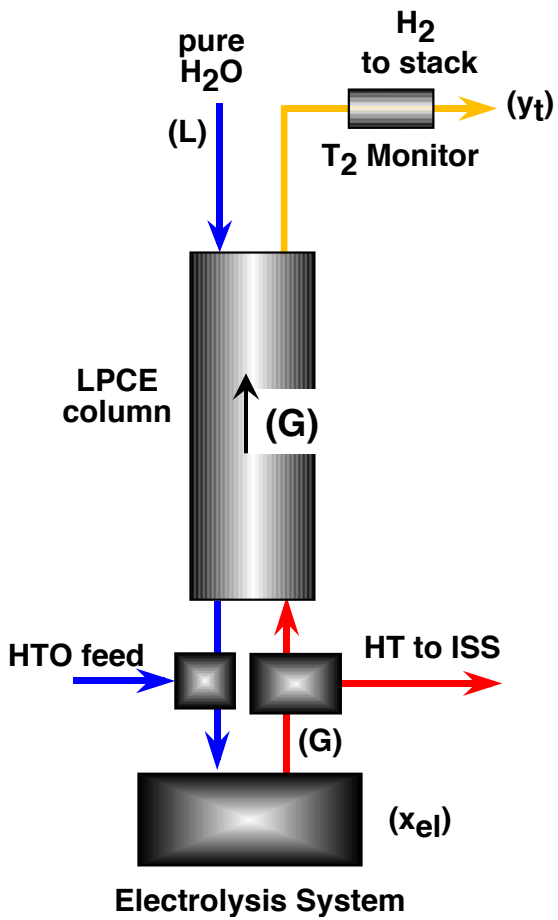
E cell Separation factor: 5 ⇨ 2

Column Sep factor: 4.9 ⇨ 1.55 @ 60 °C

Column height: ~ HETP*ln(α x_{el}/y_t)

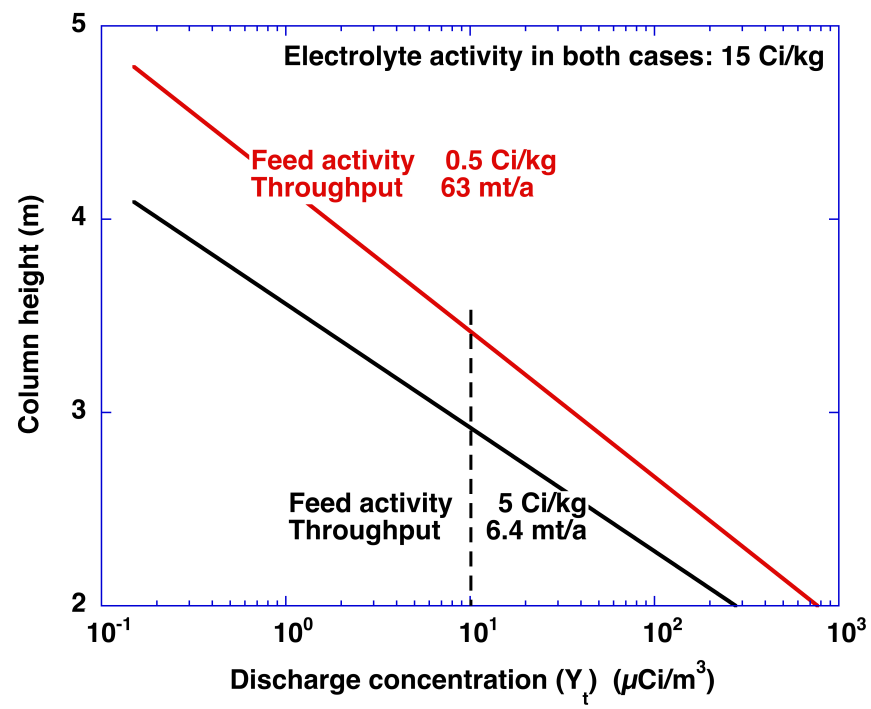


The open loop configuration provides an extra degree of freedom when processing HTO.

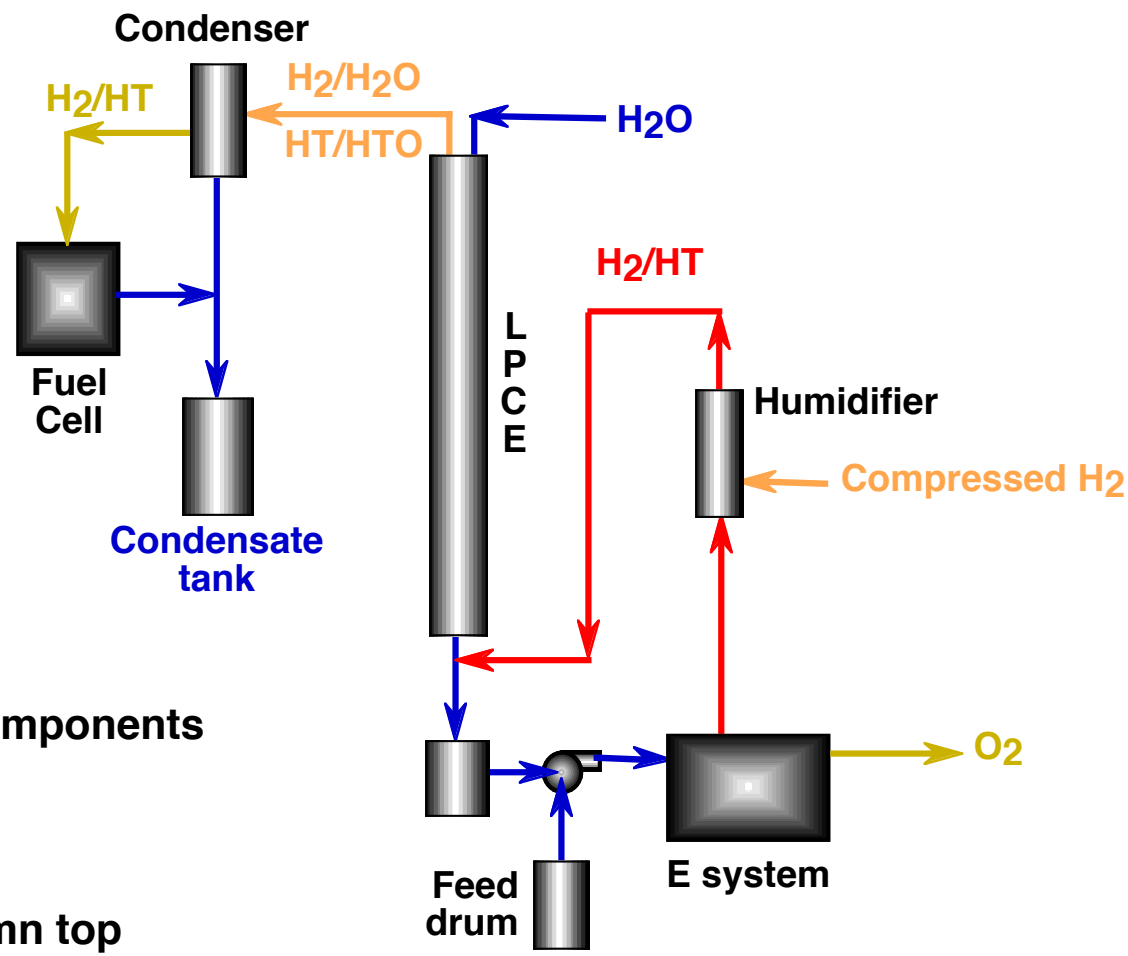


Inert rings promote: $HT + H_2O_v \rightarrow H_2 + HTO_v$

Catalyst promotes: $HTO_v + H_2O_L \rightarrow H_2O + HTO_L$



The existing CECE system has been modified to operate in an open-loop configuration up to 50 mole/s/m^2 and at variable gas-to-liquid molar ratios from 1:1 to 4:1



H_2 Flow rate:

- 75 – 500 sLPM

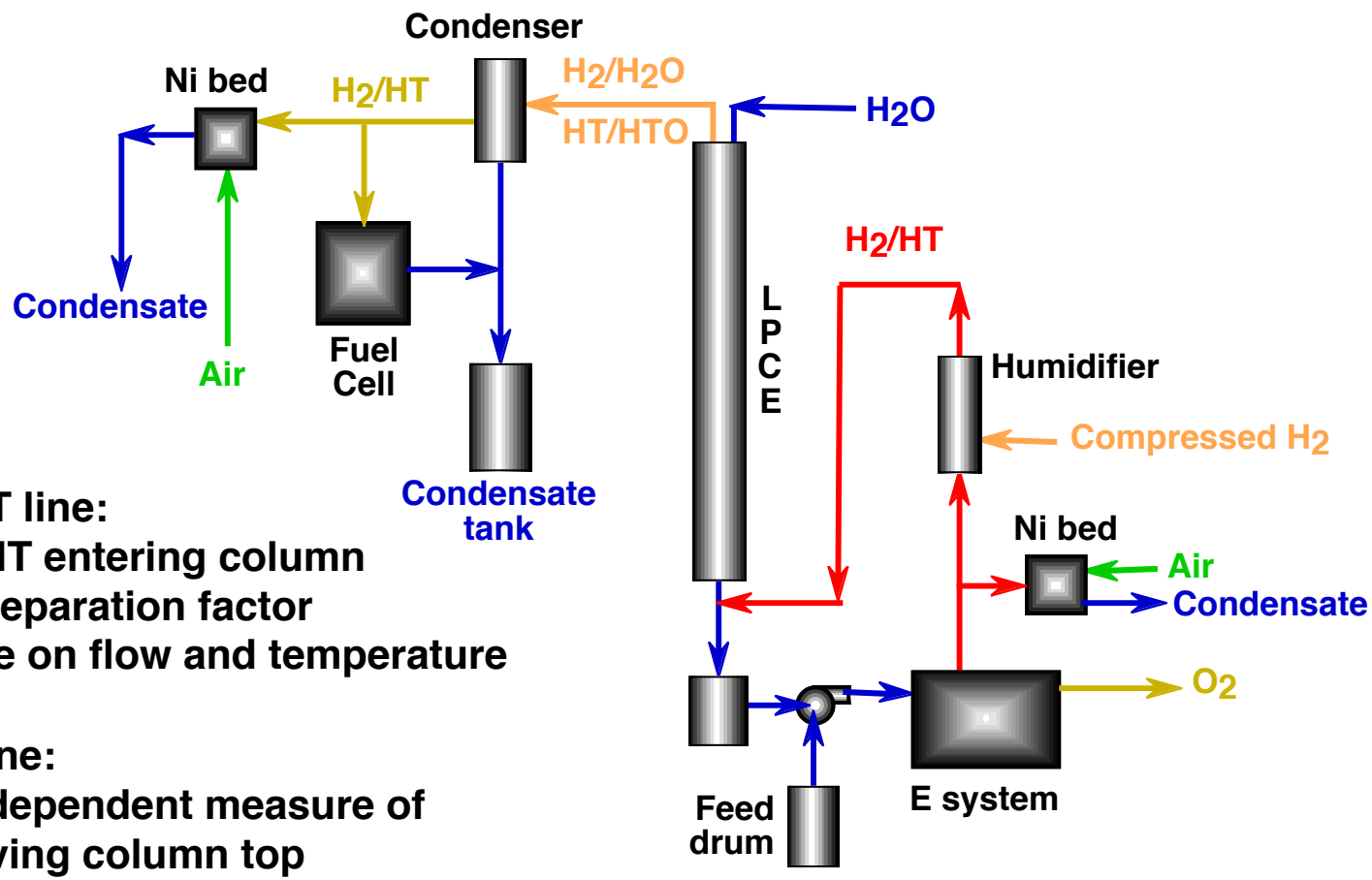
Condenser:

- Separate HTO/HT components

30 kW Fuel cell:

- Replace recombiner
- Measure HT at column top
- Energy recovery

Nickel beds measure the 'HT' activity of the hydrogen stream entering and leaving the LPCE column



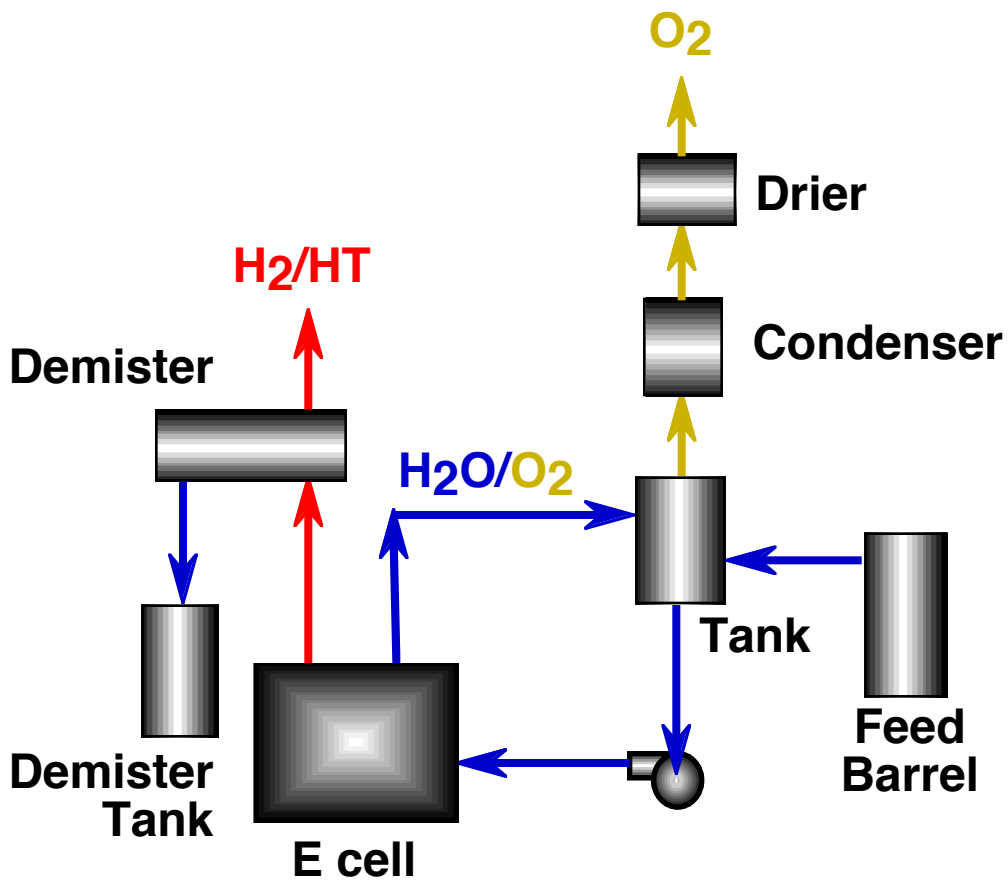
On E Cell H_2/HT line:

- Measures HT entering column
- Measures separation factor dependence on flow and temperature

On Fuel Cell Line:

- Second, independent measure of activity leaving column top

Tritiated water management is an important component in the operation of Polymer Electrolyte Membrane (PEM) electrolysis cells



Background precludes the measurement of the column performance but degraded performance is observed when the λ exceeds 4

Run #	Ecell Activity (MBq/L)	Gas to Liquid Molar Ratio (λ)	Detritiation Factor HTO	Detritiation Factor HT	HTO/HT Ratio
1	12	2.3	3200	3180	1.0
2	17	3.1	7000	3360	2.1
3	20	4.2	4600	1960	2.3
4	102	2.6	52300	14600	3.6

Gas molar flux in column: 7.8 mol/s/m^2 (75 sLPM)
Column length: 9 meter

*HT detritiation factor based on estimated activity leaving the Ecell
† For equilibrium the HTO/HT ratio ~ 5 to 6

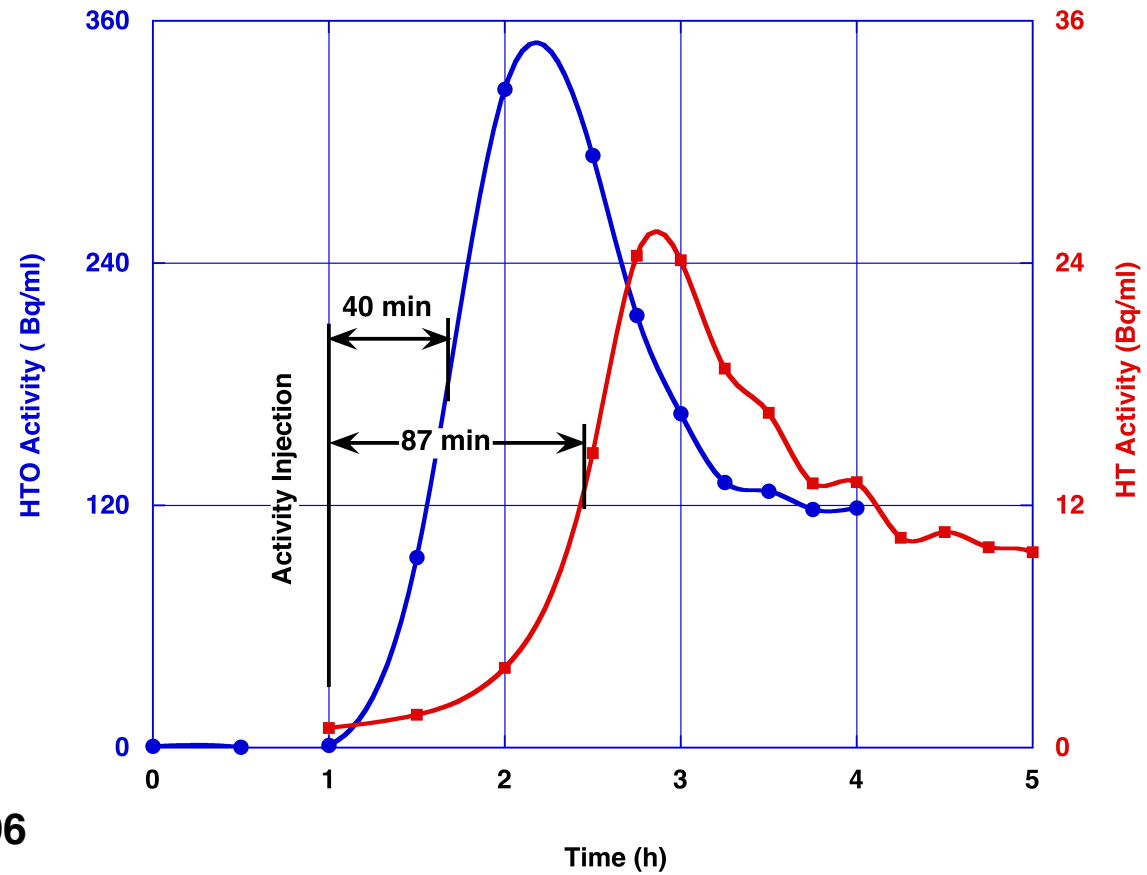
Increasing the molar flux from 7.8 to 20.9 mol/s/m^2 does not degrade the column performance.

Run #	Ecell Activity	Gas to Liquid Molar Ratio	Detritiation Factor		HTO/HT Ratio
	(MBq/L)	(λ)	HTO	HT	
1	8	2.0	9200	2520	3.7
2	13	3.0	9200	2940	3.1
3	40	1.4	12300	8500	1.4
4	50	2.0	51900	10960	4.7
5	101	2.9	66800	11130	6.0

Gas molar flux in column: 20.9 mol/s/m^2 (200 sLPM)
Column length: 9 meter

The HT activity entering the column must exceed ~ 10 MBq/m^3 to measure the actual column DF.

The columns with hydrogen flowing at 20.9 mole/s/m^{2}respond to a step perturbation in the Ecell within 30 min



λ = 2.96

The strategy to increase production capability will be staged: first optimize the process and then evaluate upgrades

- **Process optimization using the 10 cm dia column:**
  - **Increase the electrolyte activity to increase discharge above background**
  - **Measure DF as a function of G/L from 1:1 to 4:1**
  - **Measure DF dependence on G from 10 to 60 mol/s/m^2**
- **Upgrade electrolyzer from 7 sm^3/h to 100 sm^3/h:**
  - **Verify performance at high G**
- **Increase column diameter from 10 cm to 30 cm**
  - **Measure column performance with 100 sm^3/h electrolyzer**