

# Tritium Extraction from Water

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Tritiated water production is ubiquitous in facilities that handle tritium gas. Sources range from decontamination efforts, to the deliberate conversion of elemental tritium, to tritiated water in processes that strive to reduce emissions to the environment, to gaseous effluents, to the environment. At low concentrations, ranging from a few  $\mu\text{Ci/L}$  to  $\text{mCi/L}$ , high throughputs are required to process the high-volume, low-activity water. Combined electrolysis and catalytic exchange (CECE) shows promise by offering high throughput, reliability, economic viability, and facile coupling to isotopic separation systems, if necessary. This summary will discuss the features of a CECE facility based on a  $7\text{-m}^3/\text{h}$  throughput alkaline electrolysis cell.

As of 2018, there are 451 nuclear reactors operating in 31 countries with an additional 59 reactors currently under construction. All of these nuclear power stations have chronic releases of tritium that can be measured in the surrounding groundwater. In the U.S., for example, concentrations between  $20\text{ nCi/L}$  and  $0.1\ \mu\text{Ci/L}$  have been observed. The dose to an individual drinking  $4.4\text{ L}$  of  $1\ \mu\text{Ci/L}$  of water every day for a year is equivalent to only 30% of the annual dose received from natural background radiation. While the scientific community understands that the chronic release of tritium to the groundwater is not dose relevant, the public is far more sensitive to the issue. Even in locations where the ground water activity is below the EPA maximum contaminant level of  $4\text{ mrem per year}$ , land owners have successfully sued nuclear power stations over the contamination. Therefore, chronic tritium release to the groundwater remains an imminent concern for any operator of a nuclear power station. Newly constructed fission or fusion plants require robust strategies to mitigate the release of tritium to the environment to help alleviate public backlash and limit legal liabilities.

The CECE technology has been under development for several years in national laboratories<sup>1-6</sup> but has seen limited commercial deployment. The process provides an elegant, compact, and powerful option to concentrate tritium activities in water. The system comprises a liquid phase catalytic column (LPCE) integrated with an electrolysis cell. A schematic of the system is seen in Fig. 1. The electrolysis cell provides a constant supply of elemental tritiated gas to the bottom of the LPCE column. As the gas is directed up the column, a counter flow of clean water is added to the top of the column. The isotopic exchange of the tritium from the rising elemental gas to the descending water has the effect of “washing” the tritium to the bottom of the column. Clean hydrogen gas is emitted from the top of the column, while the majority of the tritium is contained in the water at the bottom. The tritiated feed stock water can be combined with the tritiated water leaving the column to collect in a tank. This water is used to feed the electrolysis cell to create more elemental gas. If the molar feed rate of the clean water at the top of the column and the tritiated feed stock water at the bottom of the column are equal to the molar rate of release of clean hydrogen from the top of the column, the system will remain in balance with all of the tritium concentrating in the water in the bottom tank.

It can be shown that the height of a packed column depends on the isotopic separation factor in the electrolyzer ( $\alpha_{\text{el}}$ ), the isotopic separation factor ( $\alpha_{\text{col}}$ ) in the column, and the concentration at the top of the column ( $y_1$ );  $\lambda$  is the ratio of the molar gas

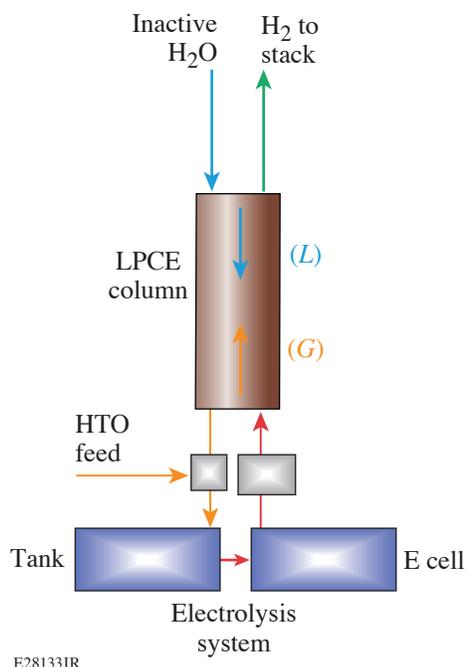


Figure 1  
Schematic of the combined electrolysis and catalytic exchange system.

flow up the column ( $G$ ) to the molar flow rate of water down the column ( $L$ ) ( $\lambda = G/L$ ); and the height-equivalent theoretical plate (HETP) for the catalyst according to the relation<sup>7</sup>

$$h = \text{HETP} * \frac{\ln\left(\frac{x_{\text{el}}}{\alpha_{\text{el}} * y_{\text{t}}} * \frac{\alpha_{\text{col}} - \lambda}{\alpha_{\text{col}} - 1}\right)}{\ln\left(\frac{\alpha_{\text{col}}}{\lambda}\right)}. \quad (1)$$

The quantities  $\alpha_{\text{el}}$ ,  $\alpha_{\text{col}}$ , and HETP are fixed when operating at a fixed temperature with a particular catalyst. For a given gas-to-liquid molar ratio, the height of a column is determined by two parameters: the concentration of the electrolyte,  $x_{\text{el}}$ , and the concentration of the effluent at the top of the column,  $y_{\text{t}}$ . Both parameters are at the discretion of the end user.

Figure 2 illustrates the column-height dependence on those two parameters for fixed  $\lambda$ . It is more economical to operate at a higher  $\lambda$  because there will be less counter-flowing water that must be electrolyzed back into elemental gas. For a chosen column height, the trade-off will be to operate at the highest possible  $\lambda$  within a prescribed emission discharge limit at the column top.

If the CECE system is being used to concentrate tritiated water for long-term storage or to reduce volume as the first step in tritium recovery, then the column height is selected for the maximum decontamination factor. Assuming the activity at the top of the column approaches zero, then in steady state, tritium balance requires that the amount of tritium introduced to the system ( $L_{\text{f}} * x_{\text{f}}$ ) must equal the amount drawn as concentrate ( $L_{\text{c}} * x_{\text{c}}$ ), where  $L_{\text{f}}$  is the molar feed rate into the electrolysis system at a concentration of  $x_{\text{f}}$  and  $L_{\text{c}}$  is the rate at which the concentrate is extracted from the system at a concentrate of  $x_{\text{c}}$ . It follows that the rate of concentrate draw is inversely proportional to the ratio of the feed activity to the concentrate activity:

$$L_{\text{c}} = \frac{x_{\text{f}}}{x_{\text{c}}} * L_{\text{f}}. \quad (2)$$

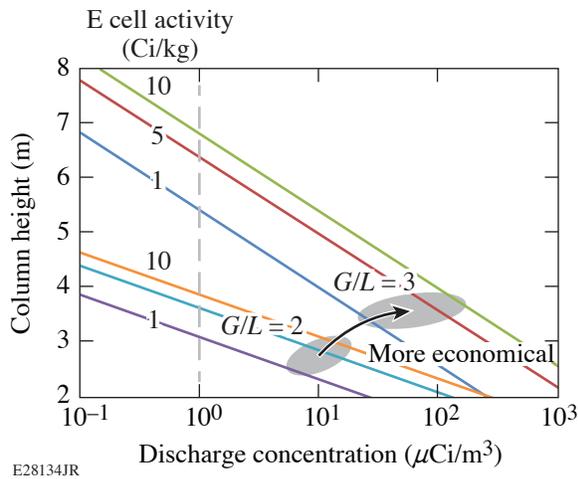


Figure 2  
Column-height dependence on the discharge concentration and the gas-to-liquid molar ratio.

As an example, assume a feed concentration of  $100 \mu\text{Ci/kg}$  is delivered to the system operating at 90% efficiency, at a gas-to-liquid molar ratio of 2 ( $\lambda = 2$ ) and driven by a  $21\text{-m}^3/\text{h}$  electrolyzer. In steady state, this system will accept 418 U.S. barrels per year (66,480 L) and produce 3.3 L of concentrate at 2 Ci/L for a volume reduction of  $\sim 20,000$ . In this example, a column height of 3.9 m would result in an emission of 0.17 Ci per year and a net activity collection efficiency of 98%. Increasing the column height by an additional 0.7 m would reduce the effluent activity by a factor of 10. At these concentrations, a portion of the electrolyzed gas can be diverted to an isotope separation system for tritium recovery.

The use of nuclear power will continue to grow in the world driven by the need for carbon neutral energy systems. Whether the reactors are fission or fusion based, it will be incumbent on the operators to reduce tritium releases to appease the public perception. CECE systems similar to one described above provide an economical and robust form of tritium concentration and recovery. The experience developed in an industrial environment attests to the simplicity and efficacy of the systems. Options to recover tritium from light water over a broad range of activities and throughputs using the small footprint of the CECE technology indicate that this technology will become an important effluent mitigation tool in the future.

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