Analytical Chemistry: A Journal (2023)
Clausius Scientific Press, Canada

DOI: 10.23977/analc.2023.020114
ISSN 2616-1958 Vol. 2 Num. 1

Process Tritiated Water through a Bioreactor: An Ideal Model for Dealing the Fukushima Wastewater

Leyi Li[1,2,a,*]

[1]*Beijing 101 High School, 11# Yiheyuan Road, Beijing, China*
[2]*Pioneer Research Course, Oberlin College, Oberlin, Ohio, USA*
[a]*meris0918@outlook.com*
[*]*Corresponding author*

Keywords: Tritium, Bioreactor, Detritiation

Abstract: The existing tritium management is marked by high energy and financial costs. It is dangerous to directly release such a radioactive element into the environment. To deal with this dilemma, this paper proposes a model of a bioreactor that can detritiate the wastewater depending on plants' tritium incorporation ability. Tritiated water (HTO) can be converted into organically bounded tritium (OBT) through photosynthesis and various metabolism processes and the stored in plants. Seaweeds and bryophytes can be candidates for the tritium absorber. The incorporated tritium can be processed through a combination of freeze-drying and oven-drying methods and then stored separately, decaying into the stable helium-3.

1. Introduction

Tritium is a radioactive element that occurs often in a low concentration in the environment. However, anthropogenic products, such as the tritium resulting from the use of heavy water add to the natural volume, therefore causing an increment in concentration. The uptake of contaminated chemicals should be considered a danger since a proportion of intake will stay in an organic form, remain in the human body, and thus emit rays externally. Currently, several studies suggest methods for detritiation, but systems share problems such as high energy use; high cost; and low security.

1.1. Prerequisites for tritium

Tritium (^3H or T) is a rare isotope of hydrogen with a half-life of 12.31 years, and it is radioactive, emitting low-energy beta particles and finally decaying into helium-3, a stable isotope[1][2][3]. The emitted beta particles are of such low energy that the skin blocks the radiation from doing harm. In the environment, tritium exists in different forms, and scientists mainly delve into three of them: tritiated gas (HT), tritiated water (HTO), and organically bounded tritium (OBT)[4]. Even though tritium is produced naturally in the atmosphere through reactions between a fast neutron and atmospheric nitrogen[5], human activities such as the use of nuclear weapons and accidental leakage from nuclear powerplants significantly contribute to the tritium volume in the atmosphere and biosphere. Moreover, the artificial products break the biogeochemical equilibrium[6], causing unpredictable damage on Earth.

1.2. Consequences of man-made release

The arguments around “safe discharge” remain contested. It is indeed a low-energy beta emitter and will disappears within a few decades by radioactive decay; however, the threat to humans and the environment needs to be weighed in the long run. Considering the behavior of tritium, Eyrolle-Boyer et al., (2014) point out that the “free” forms (i.e. HT, HTO, and tissue-free water tritium (TFWT)) are closely linked to the water cycle and carbon cycle[2], which means that this radioactive element can be transported in the entire environment through evaporation, precipitation, and the waterway above and beneath the surface. In addition, organisms will incorporate it with organic compounds, forming OBT during the metabolic process. OBT is much more dangerous to humans. Instead of exposing to tritium externally, OBT will remain internally and thus emit beta rays directly to inner tissues that are not sheltered by barriers like the skin. Though HTO cannot bioaccumulate in the environment, whether OBT has the same property is still unclear[7], and the bioaccumulation and biomagnification processes seem plausible based on current knowledge. After HTO is taken in, it bounds to carbon molecules to form the nonexchangeable OBT (NE-OBT) or the other molecules like nitrogen to form exchangeable OBT (E-OBT). Moreover, NE-OBT, behaving as its name suggested, can retain much longer in tissues compared to the E-OBT due to the stronger bounding[8]. As a result, the OBT form is not only like a buried time bomb, having the probability of causing genetic and cytogenetic impacts, but also might amass along the food chain.

In this case, though the International Atomic Energy Agency (IAEA) “confidently” confirmed that “the international environmental protections objectives will be amply met by the controls in place for the discharge of [Advanced Liquid Processing System (ALPS)] treated water and that the dose rates to biota are negligible compared to the international safety criteria set by [International Commission on Radiological Protection (ICRP)]”[9], the Japanese government should be more farsighted and decisively cancels the discharge plan. Tokyo Electric Power Company (TEPCO) insists on releasing the water contaminated by the meltdown of Fukushima Daiichi nuclear power plant into the Pacific Ocean while claiming that the wastewater will no longer be harmful to the environment after being processed by an advanced system and further diluted by seawater[10]. However, the belief, “The solution to pollution is dilution,” has been proven to be unfeasible. Zhao et al. (2021) built a model for the dispersion of tritium released from the Fukushima Daiichi nuclear plant and found that the spreading area extended both horizontally and vertically[11]; moreover, the tritium concentration in sea surface water maintained at a certain concentration for a prolonged period which strongly refutes the idea of solving the problem through dilution.

1.3. Current processing system

Laboratories around the world have researched detritiation methods for decades. Presently, there are several techniques for processing tritiated water, and the combined electrolysis catalytic exchange (CECE) and the liquid phase catalytic exchange (LPCE) are two effective and promising methods that can be operated under a milder condition in comparison to the other, like vapor phase catalytic exchange (VPCE), which requires high temperature and pressure[12][13]. Nevertheless, among detritiation facilities, none of them can truly balance these applicative factors: energy use, efficiency, waste disposal, and cost[14]. Accordingly, this paper is going to suggest a model of a bioreactor that aims to be an economical plan and achieve nearly zero net energy use, and less waste disposal while still having a comparatively high removing effect. This environmentally friendly design will be an ideal substitute for conventional systems and can reduce many safety concerns.

2. An ideal model of the bioreactor

The bioreactor operates depending on the TFWT-OBT conversion process. Plants take in HTO while absorbing water to survive. They perform photosynthesis, a process that creates organic materials from CO2 and the tritiated water, and other metabolic processes incorporating tritium, therefore reducing the tritium in the wastewater tank over time, achieving the detritiation process without arduous efforts. To build this system, two questions should be answered: how tritium moves and is held up within plants, and what plant species is the best candidate playing as an OBT holder.

2.1. Tritium's movement between plants and the environment

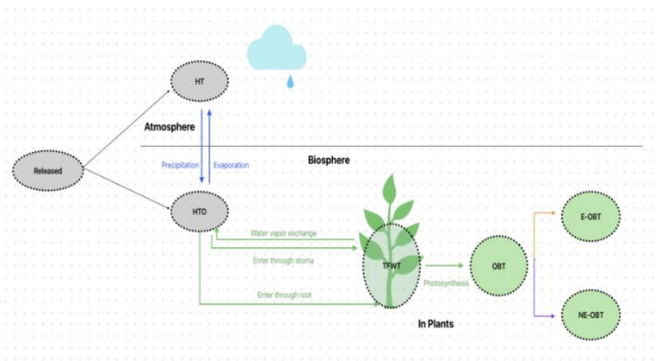

Figure 1: Tritium's movement between the atmosphere and plants.

According to Figure 1, plants generally uptake tritium in two ways as they present in the water cycle: directly transfer tritium into stomas on the foliage or through their roots[2]. The form of tritium is HTO, which can be the oxidated insoluble HT or the contaminated water release[15]. Accompanying the water circulation, TFWT, namely the mobile form of tritium after entering an organism, freely diffuses to different organs of the plant and a fraction of them are involved in photosynthesis and various metabolism processes, converting into OBT[16].

2.2. Plant selection for designing the bioreactor

Since the mechanism of this bioreactor is tritium incorporation, keeping internal tritium from re-release is the goal of the design. Considering the trait of tritium, selected plant species should be able to eliminate the following two concerns.

Firstly, both forms of OBT, E-OBT and NE-OBT, are strongly attached to the organism; however, E-OBT contains tritium atoms that are easily exchanged with hydrogen atoms in water molecules due to the weaker bonds, which means that only NE-OBT has prolonged retention[17][18].

Secondly, since the formation of OBT results from the metabolic process, the OBT conversion rate correlates with the plant's metabolism capability. However, OBT loss is inevitable due to the fundamental demand for organic materials[19].

As a result, a desirable species should absorb water rapidly; have a moderate rate of metabolism; and have a simple life cycle and anatomy since plants incorporate OBT at different rates at each growing phase and in different organs, which will make the system operation complicated[5].

2.2.1. Seaweed

Seaweed, or macroalgae, refers to a wide range of marine organisms living in the photic region[20]. They generally consist of the lamina, a leaf-like structure, and the holdfast, a root-like structure. The serried lamina offers a greater surface area that allow them to be exposed to enough sunlight[20]. Mao et al., (2023) gave specific data that in a species called *D. salina*, the intrinsic percentage of carbohydrates, proteins, and lipids are 54.3%, 30.6%, and 15.1% respectively[21]. This high protein and carbohydrate proportion suggests that seaweed might have a competitive OBT conversion rate while also keeping it in an immobile form in proteins, or that a small fraction of lipids. Studies measuring the influence of released tritiated water commonly used algae as a test object, and they all tested OBT in their algae samples[16][22]. Additionally, the measured OBT concentration in the algae was much higher than in the ambient water, which refers to the incorporation and accumulation process.

If seaweed is selected to be the tritium absorber in the bioreactor, careful selection of species and maintenance would boost the efficacy. Firstly, since seaweed species are diverse in their color and lamina size, these differences related to the photosynthetic rate should be tested to find the one with the highest value. Secondly, Mao et al., (2023) suggested that the seaweed in the growing phase has a faster photosynthesis rate than the group in the stationary phase, which means a faster incorporation into organic materials[21]. Furthermore, the light shielding effect causes a decrease in OBT accumulation, which suggests that the algae's growth should be monitored so that the shielding effect can be avoided by scheduling a harvest cycle.

2.2.2. Non-vascular plants

Nonvascular plants are those lacking vascular tissues, the xylem and phloem. Boyer et al., (2009) suggested nonvascular plants as a particular case of the transfer of tritium between themselves and the environment because they can absorb the ambient water quickly while are not able to control the process of water vapor exchange[5]. However, the inability to retain and deliver internal water doesn't affect the OBT retention level. Several studies mentioned the incorporation ability of bryophytes such as mosses and lichens. For example, Daillant et al. (2004) proposed the use of lichens as indicators of tritium and discovered that the OBT activity in lichens was higher than the background by a factor of 1000 and even had this advantage in a distance[23]. The authors point out that the slow metabolism equips lichens with such suitability, which also aligns with the previous assumption of the best candidate of plant species.

If bryophyte is selected to be the tritium absorber in the bioreactor, selecting the appropriate species from their miscellaneous options is still an arduous but vital step. Daillant, et al. (2004) suggested that a greater surface/mass ratio contributes to a higher OBT concentration, and the fruticose lichens (E. prunastri) they sampled may be an alternative in the future study[24]. Another concern is that since lichens grow on land, merely placing it in the tank like placing the seaweed is not feasible. As a result, using lichens as the tritium absorber requires a new well-designed device in response, while a close relative, hornwort, a type of aquatic plant, now lacks studies to compare its OBT retention level to lichens in the same area.

2.3. Tritium disposal

After a few days, the plants should accumulate a certain amount of OBT in their body and reduce the amount of tritium in the wastewater. While the detritiated water flows to the subsequent tank, according to Mao et al. (2023), plants need to be renewed periodically as the incorporation rate will decrease when plants enter their stationary phase[21]. The collected plants are filled with OBT and need to be settled properly. Considering the immobility of tritiated organic matter, dehydration is a necessary step ensuring only OBT will be collected. Moreover, the volume and weight of the plants can be largely reduced, which benefits further storage. Freeze-drying is a popular strategy used to improve the product stability through a three-step process of freezing and drying, which is commonly used in the pharmaceutical industry[25]. It is also an acknowledged method used to prepare testing samples for measuring OBT in plants. Kim, S.-B., & Roche, J. (2013) suggest that by using the freeze-drying method, an average of 98.5% of water can be removed from the vegetable samples, and an average of 99.9% of water can be removed when combining oven-drying with freeze-drying[7]. In this way, the dehydrated plants can be dumped to local landfill sites, and the tritium within will decay into helium-3, a stable isotope, as time elapses.

3. Unsolved problems

The bioreactor proposed in this paper is an ideal model, which means that its efficiency and applicability require further experiments to evaluate in light of uncertainties.

Firstly, since the efficiency is closely related to proper plant selection, a series of experiments should be conducted to figure out which plant species is suitable for dealing with tons of Fukushima wastewater. The incorporation rate should be recorded, and the length of the processing period, or the plant renewal cycle, should be settled according to the incorporation rate and OBT content remaining in plants.

Secondly, methods of boosting plants' detritiating capability need to be tested and retested. For example, whether a combination of multiple plant species can improve the detritiation rate is currently unknown and worthwhile to be verified in the experiment. Moses & Calvin (1959) discovered that the NE-OBT formation rate is three times higher under the light than in the dark but only used Chlorella cells, which is not convincing enough and requires further studying[26].

Thirdly, to measure the applicability of the bioreactor, several factors should be considered and balanced: tritium concentration of the processed water; sustainability, which refers to the storing stage; and the cost. A horizontal comparison between the bioreactor and existing strategies is also necessary, which not only serves as evidence of the priority of this all-natural system but also helps to promote an update in tritium management.

4. Application and Evaluation

Apart from the efficacy, saving energy is another goal of this bioreactor. An all-natural bioreactor is not an unexplored field. For example, Omega's Eco Machine™ is an eco-friendly system relying on plants, bacteria, algae, snails, and fungi to recycle domestic wastewater into clean water that restores the aquifer[27]. It can process a copious amount of water per day and form a closed-loop hydrological cycle within the system so that every drop of water can maximize its utilization. Their success is the reward of careful design and creativity, and it is possible to achieve the same effect on detritiation if we learn this purification equation. Referring to Omega's Eco Machine™, the prophase, solid settlement, can be omitted in this bioreactor for detritiation since the water flowing in is pretreated by Tokyo Electric Power Company's advanced machine, which tackles the heavy metals by conducting co-sedimentation, adsorption, and physical filtration but neglects C-14 and tritium[10]. Then, plants in the tank will finish their responsibility of incorporating tritium. When tritium is converted into OBT and stored in periodically replaced plants, the treated water may flow into an artificial wetland as the ordinary non-radioactive wastewater waiting for further biochemical procession. Finally, clean water can be delivered to multiple destinations, or be recollected and utilized by the nuclear processing facility itself.

How to deal with nuclear wastewater is a serious global issue. Recently, in August 2023, New York citizens protested and urged the government to sign a bill preventing the owner of the Indian Point nuclear plant from releasing tritiated water into the Hudson River. The wastewater is promised to be stored until a safe method of disposal can be determined. This legislation is a great success, but the topline is finding an effective method of processing the contaminated water while leaving fewer additional environmental damages. Only achieving this goal can allow our offspring to be free to enjoy beautiful river scenery in the future.

In this case, it is worthwhile to propose this design of a bioreactor. The whole process, except the existing resolution in prophase, minimizes energy and financial costs. Artificial wetlands at the final stage even have ornamental value. If the experimental data confirms the effectiveness of plants' incorporation, this bioreactor would be a tremendous progress and largely reduce the burden on the environment, therefore benefiting humans in the long run.

5. Conclusion

The damage resulting from tritium pollution is long-lasting and impalpable at the beginning. Though it is a weak radioactive element, the released tritiated water will accumulate in organisms and sediments in the organic form causing internal exposure to radiation. The bioreactor utilizes the tritium incorporation by plants, absorbing tritium from the wastewater and converting it into NE-OBT that can be recollected through freeze-drying. In this way, the tritium concentration can be further reduced, ideally to zero while the extracted NE-OBT will be stored separately and decay in decades. If this invention is confirmed to be effective, the Fukushima wastewater problem may be solved economically and in an environmentally friendly way.

Acknowledgement

Throughout the writing of this paper I have received a great deal of support and assistance.

I would first like to thank my Professor, Stuart Belli, whose expertise was invaluable in facilitating the research questions and methodology. You always encouraged me to delve deeper and think broader. Your insightful feedback pushed me to sharpen my thinking and brought my work to a higher level.

I would also like to thank my cohort advisor, Damian Emory, who was always there for my technical problems and helped me to solve scheduling conflicts.

In addition, I would like to thank my tutors from the Pioneer Writing Center, Eleda Fernald, Evie Ford, and Liana Hakobyan, for their valuable guidance on academic language using and writing format.

Finally, I would like to thank Pioneer Academics for providing me a wonderful opportunity to conduct my own research, improve my academic rigor, and discuss with insightful young scholars around the world.

References

[1] Eyrolle, F., Ducros, L., Dizès, S. L., Beaugelin-Seiller, K., Charmasson, S., Boyer, P., & Cossonnet, C. (2018). An updated review on tritium in the environment. Journal of Environmental Radioactivity, 181, 128–137. https://doi.org/ 10. 1016/j.jenvrad.2017.11.001

[2] Eyrolle-Boyer, F., Boyer, P., Claval, D., Charmasson, S., & Cossonnet, C. (2014). Apparent enrichment of organically bound tritium in rivers explained by the heritage of our past. Journal of Environmental Radioactivity, 136, 162–168.

[3] Zhao, C., Wang, G., Zhang, M., Wang, G., Bezhenar, R., Maderich, V., Xia, C., Zhao, B., Jung, K. T., Periáñez, R., Akhir, M. F., Sangmanee, C., & Qiao, F. (2021). Transport and dispersion of tritium from the radioactive water of the Fukushima Daiichi nuclear plant. Marine Pollution Bulletin, 169, 112515. https://doi.org/10. 1016/j. marpolbul.2021.112515

[4] Kim, S., Baglan, N., & Davis, P. A. (2013). Current understanding of organically bound tritium (OBT) in the environment. Journal of Environmental Radioactivity, 126, 83–91. https://doi.org/10.1016/j.jenvrad.2013.07.011

[5] Boyer, C., Vichot, L., Fromm, M., Losset, Y., Tatin-Froux, F., Guétat, P., & Badot, P. (2009). Tritium in plants: A review of current knowledge. Environmental and Experimental Botany, 67(1), 34–51. https://doi.org/ 10. 1016/j. envexpbot.2009.06.008

[6] Eyrolle, F., Copard, Y., Lepage, H. et al. Evidence for tritium persistence as organically bound forms in river

sediments since the past nuclear weapon tests. Sci Rep 9, 11487 (2019). https://doi.org/10.1038/s41598-019-47821-1
[7] Kim, S.-B., & Roche, J. (2013). Empirical insights and considerations for the OBT inter-laboratory comparison of environmental samples. Journal of Environmental Radioactivity, 122, 79–85. doi: 10.1016/j.jenvrad.2013.03
[8] Melintescu, A., & Galeriu, D. (2011). Dynamic model for tritium transfer in an aquatic food chain. Radiation and Environmental Biophysics, 50(3), 459–473. https://doi.org/10.1007/s00411-011-0362-0
[9] IAEA. (2023). IAEA Comprehensive Report on the Safety Review of the Alps-treated Water at the Fukushima Daiichi Nuclear Power Station. International Atomic Energy Agency https://www.iaea.org/sites/ default/files/ iaea_ comprehensive_alps_report.pdf
[10] Nogrady, B. (2023). Is Fukushima wastewater release safe? What the science says. Nature, 618(7967), 894–895. https://doi.org/10.1038/d41586-023-02057-y
[11] Zhao, C., Wang, G., Zhang, M., Wang, G., Bezhenar, R., Maderich, V., Xia, C., Zhao, B., Jung, K. T., Periáñez, R., Akhir, M. F., Sangmanee, C., & Qiao, F. (2021). Transport and dispersion of tritium from the radioactive water of the Fukushima Daiichi nuclear plant. Marine Pollution Bulletin, 169, 112515. https://doi.org/ 10. 1016/j. marpolbul.2021.112515
[12] Boniface, H., Gnanapragasam, N. V., Ryland, D., Suppiah, S., & Perevezentsev, A. (2017). Water Detritiation System for ITER—Evaluation of design parameters. Fusion Science and Technology, 71(3), 241–245. https://doi.org/ 10.1080/ 15361055.2017.1290970
[13] Fu, Q., Feng, X., Yin, X., Song, Y., & Xu, Y. (2021). A highly efficient and stable catalyst for liquid phase catalytic exchange of hydrogen isotopes in a microchannel reactor. International Journal of Hydrogen Energy, 46(43), 22446–22453. https://doi.org/10.1016/j.ijhydene.2021.04.073
[14] Wu Z.; Hu S.. Tritium Removal from Tritiated Heavy Water[J]. Journal of Isotopes, 2021, 34(1): 89-95.
[15] Belot, Y., 1986. Tritium in plants: a review. Radiat. Prot. Dosim. 16 (1–2), 101–105.
[16] Baburajan, A., Sudheendran, V., Gaikwad, R. H., Ravi, P., Nayak, R., D'Souza, S. R., & Karunakara, N. (2019). Tissue free water tritium (TFWT) and organically bound tritium (OBT) in marine eco system at Tarapur on the west coast of India. Journal of Radioanalytical and Nuclear Chemistry, 323(3), 1431–1441. https://doi.org/10.1007/s10967-019-06861-7
[17] Kim, S. B., Workman, W., & Davis, P. A. (2008). Experimental investigation of buried tritium in plant and animal tissues. Fusion Science and Technology, 54(1), 257–260. https://doi.org/10.13182/fst08-a1807
[18] Kim, S., & Korolevych, V. (2013). Quantification of exchangeable and non-exchangeable organically bound tritium (OBT) in vegetation. Journal of Environmental Radioactivity, 118, 9–14. https://doi.org/10.1016/j.jenvrad.2012.11.006
[19] Diabaté, S., & Strack, S. (1997). Organically bound tritium in wheat after short-term exposure to atmospheric tritium under laboratory conditions. Journal of Environmental Radioactivity, 36(2–3), 157–175. https://doi.org/10.1016/s0265-931x(97)84985-5
[20] Mišurcová, L. (2011). Chemical Composition of Seaweeds. Handbook of Marine Macroalgae, 171–192. https://doi.org/10.1002/9781119977087.ch7
[21] Mao, L., Lu, K., Yu, M., Dong, S., Cui, F., Zhang, Y., Diwu, J., Chai, Z., & Wang, S. (2023). Algae induced tritium organification promotes human exposure risk. Research Square (Research Square). https://doi.org/10.21203/rs.3.rs-3256924/v1
[22] Bonotto, S., Arapis, G., Mellet, J., Hoursiangou-Neubrun, D., Cinelli, F., Belcari, P., Gerber, G. B., & Kirchmann, R. (1984). Behavior of tritium in the marine environment: Uptake and distribution of tritiated water and of tritiated organic compounds in unicellular and multicellular algae. Hydrobiologia, 116–117(1), 187–189. https://doi.org/ 10.1007/bf00027662
[23] Daillant, O., Kirchner, G., Pigrée, G., & Porstendörfer, J. (2004). Lichens as indicators of tritium and radiocarbon contamination. Science of the Total Environment, 323(1–3), 253–262. https://doi.org/10.1016/j.scitotenv.2003.09.030
[24] Daillant, O., Boilley, D., Gerzabek, M. H., Porstendörfer, J., & Tesch, R. (2004). Metabolised tritium and radiocarbon in lichens and their use as biomonitors. Journal of Atmospheric Chemistry, 49(1–3), 329–341. https://doi.org/10.1007/s10874-004-1245-4
[25] Abla, K. K., & Mehanna, M. M. (2022). Freeze-drying: A flourishing strategy to fabricate stable pharmaceutical and biological products. International Journal of Pharmaceutics, 628, 122233. https://doi.org/ 10. 1016/j. ijpharm.2022.122233
[26] Moses, V., & Calvin, M. (1959). Photosynthesis studies with tritiated water. Biochimica Et Biophysica Acta, 33(2), 297–312. https://doi.org/10.1016/0006-3002(59)90117-9
[27] Omega Institute. (2021). The eco machine. eomega.org. https://www.eomega.org/center-sustainable-living/eco-machine