Radioactivity Analysis of Tritium Contaminated Wood Sample by Electrolysis of Tritiated Water and Extraction by Combustion Method

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1. Introduction

In working place deal with radioactive nuclide, the material and environment around working place can be contaminated by radioactive nuclide. Those material should be classified radioactive waste and disposed after radioactivity analysis. Because tritium cause ionization damage in human body [1], it is hazardous when it enters human body by respiration or intake. Tritium contamination of wood sample according to gaseous tritium (HT) generated on various electrolysis condition and extraction by combustion were researched in this study.

2. Methods

For generation of gaseous tritium, electrolysis system including proton exchange membrane (PEM) cell was used. For extraction of tritium from wood sample, high temperature furnace (Pyrolyser, Raddec International Ltd) was used.

2.1 Electrolysis of Tritiated Water

When distilled water (only containing H_2O molecules) is entered to PEM cell, water molecules are decomposed in hydrogen and oxygen gas by electrolysis. When tritiated water including tritium molecules (HTO) is electrolyzed, gaseous tritium (HT) and hydrogen gas are generated. Distilled water circulates from inlet of PEM cell to outlet of PEM cell. Distilled water losses electrons in PEM cell. Water molecules lose electron in the anode of PEM cell in this circulation process. And then, hydrogen ions generated by loss of electrons are moved from anode to cathode through PEM. Electrons are moved to cathode through electrical circuit in PEM cell. As shown in Fig. 1, hydrogen gas is generated by unite of hydrogen ions and electrons on the cathode of PEM cell.

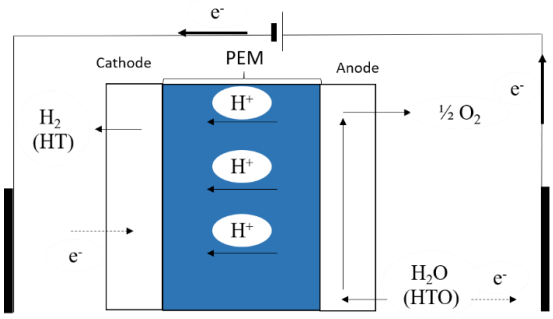

Fig. 1. The schematic diagram of electrolysis using PEM cell

Using same principle, electrolysis of tritiated water can generate gaseous tritium. Water circulates from water container to PEM cell by moving force of water pump. Small amount of the tritiated water that is not electrolyzed. And it was also moved to water trap together with gaseous tritium. However, there is pipe connected to outlet on upper side of water trap. Therefore, small amount of the tritiated water was confined to water trap by gravity, and only gaseous tritium can be moved to outlet. As shown Fig. 2, wood samples (10 mm x 7 mm x 50 mm) in the water trap are contacted to gaseous tritium flow and contaminated by ion exchange reaction [2].

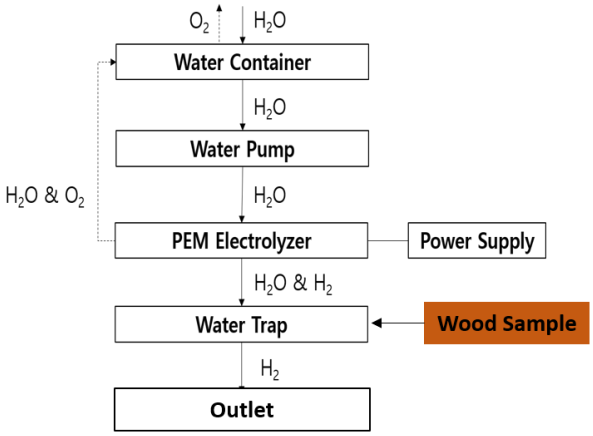

Fig. 2. The schematic diagram of gas flow in electrolysis system

2.2 Contamination of Wood Sample

From tritiated water, the amount of gaseous tritium by electrolysis can be different as electrolysis conditions. Electrolysis duration, current supplied to PEM cell from power supply, radioactivity concentration of tritiated water are used as variables in this study. If the amount of gaseous tritium generated by electrolysis is changed, the degree of contamination of wood sample can become different. Table I shows electrolysis experimental conditions on experiments with six wood samples. From sample 1 to sample 3, radioactivity concentration of tritiated water used to electrolysis is same, and radioactivity concentration of tritiated water used to experiments using sample 4 to sample 6 is double of it. The product of time and current, 8 is all same in six experiments. Mass flow rate of gaseous tritium generated

by electrolysis is proportional to current. Mass flow rate per 1 ampere is 70 cc/min.

Table I: Electrolysis experimental conditions

Wood sample number	1	2	3	4	5	6
Radioactivity concentration of tritiated water (kBq/L)	3680.28			7360.56		
Electrolysis duration (hours)	4	2	1	2	1	0.5
Current (A)	2	4	8	2	4	8

2.3 Combustion method

After electrolysis experiment, contaminated wood sample is shipped to sample boat of high temperature furnace. The inner temperature of device was set to 120℃. And air and oxygen gas flows from quartz tube including sample boat shipped wood sample. By flow of gases, tritiated water molecules inside of wood sample becomes tritiated water vapor and separated from wood sample. Separated tritiated water vapor are captured to 0.1 M nitric acid solution connected to end of quartz tube. After drying for 11 hours, wood sample is combusted on temperature of 500 ℃. It is process to extract residual tritiated water from wood sample by combustion. In this process, wood sample should be decomposed to fine piece physically for protection of incomplete combustion as Fig. 3. Nitric acid solution is mixed with liquid scintillator (Gold star) for making liquid scintillation counter (LSC) cocktail. Mixing volume ratio between nitric acid solution and liquid scintillator was 8:12.

Fig. 3. The wood sample by incomplete combustion

3. Results

Table II shows radioactivity concentration of the tritium extracted from wood samples.

Table II: Radioactivity concentration of extracted tritium from wood sample

Wood sample number	Radioactivity concentration of extracted tritium from wood sample (Bq/g)	Minimum detectable activity (Bq/g)
1	24.28±0.37	0.02564
2	34.09±0.46	0.02999
3	34.01±0.40	0.02619
4	23.23±0.33	0.03121
5	26.18±0.39	0.02164
6	26.07±0.36	0.02258

From wood sample 4 to 6, radioactivity concentration of tritiated water was twice of experiments using wood sample 1,2 and 3. However, electrolysis durations of wood sample 1,2 and 3 were higher than those of sample 4,5 and 6. Theoretically, the results about extracted tritium from 6 experiments were expected to be similar. However, they were all different. In case of experiment with same electrolysis current, the radioactivity concentration of tritium from wood sample 1 is higher than that of wood sample 4. When wood sample 2 and 3 were compared to wood sample 5 and 6, results from 2 and 3 were higher than those from 5 and 6. When tritiated water with same radioactivity concentration is electrolyzed, radioactivity concentration of extracted tritium was highest in wood sample 2 and 5.

4. Conclusions

Tritium contamination of wood sample can be caused by gaseous tritium (HT) as well as tritiated water (HTO) or tritiated water vapor. And then, when wood sample is contaminated by gaseous tritium from electrolysis using PEM cell, electrolysis duration dominates more than radioactivity concentration of tritiated water. However, in case of radioactivity concentration of tritiated water used for electrolysis is same, higher mass flow meter caused by higher current dominate more than electrolysis duration. This study can be used as the basis of the research about contamination of wood sample in various environment existing tritium.

REFERENCES

[1] A. M. Soreefan • T. A. DeVol, Determination of Tritium Enrichment Parameters of A Commercially available PEM Electrolyzer: a Comparison with Conventional Enrichment Electrolysis, Journal of Radioanalytical and Nuclear Chemistry, 282, p 513, 2009.
[2] M. Nishikawa, N. Nakashio, T. Shiraishi, S. Odoi, T. Takeishi, K. Kamimae, Tritium Trapping Capacity on Metal Surface, Journal of Nuclear Materials, pp. 99-105, 2000.