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The Separation of Tritium Radionuclide from Environmental Samples by Distillation Technique

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1. Introduction

Tritium is an isotope of hydrogen which emit beta particles in energy range of 5 – 18.6 keV, and has relatively long half life of 12.3 years. In the environment, tritium is originated both from the nature and human activities. Naturally, tritium is produced in the atmosphere by interaction of cosmic rays with the nuclei of atoms present in the air, and estimated to contribute the annual effective dose equivalent about 0.01 μSv (United Nation, 1988). The other natural sources of tritium are nuclear reactions in earth’s crust and environmental materials.

The sources of tritium in the environment relating to human activities, includes atmospheric nuclear testing, continuous tritium release from nuclear reactor under normal operation or accident, and incidental release from consumer products such as paint, watch materials, and other fluorescence goods (Glastone & Jordan, 1980; Puhankainen & Heikkinen, 2007). The food, drink and the use of such products can cause tritium contamination to the human body (Konig, 1990) and contribute the internal radiation doses to the tissues since tritium is distributed to the whole body (Trivedi et al., 2000).

As tritium in the environment possibly contaminates humans, the measurement of tritium content in environmental materials as well as in human body samples is needed to be conducted to evaluate the tritium β radiation doses probably received by the body. Unfortunately, β radiation from tritium is not so simple to be determined without sample preparation for separating tritium from the sample bulk. Since tritium is a hydrogen isotope, it is possible to take tritium out from the samples as tritiated water molecule lead to an easy measurement using liquid scintillation counting (LSC) method.

Distillation technique has been chosen as a simplest technique to be used in tritium separation from the environmental samples. Some distillation techniques have been developed to take the tritium out from various environmental material including air, water, soil, plants, and biological materials such as human urine.

This chapter describes the developed distillation technique applied in environmental tritium analysis. The performance of the developed distillation techniques in tritium separation from the environmental samples are discussed based on the experimental test.
1.1 Environmental tritium

The majority of tritium in the environment, both naturally and anthropogenic, is present in the form of water molecule (named HTO), while the others are gas (named HT and CH$_3$T) and organic molecules. As an isotope of hydrogen, tritium can easily enter various environmental materials containing hydrogen. In the environmental component tritium is bonded to water molecule, called free-water tritium (FWT) and also to the organic compound as organically bound tritium (OBT).

Tritium in the environmental material is possible to contaminate human body through the food chain. The drinking water, food, inhaled air, and skin absorption are the main sources of tritium for human body contamination. Tritium is uniformly distributed in the body liquid 2 until 4 hours after its inhalation, ingestion or absorption through the skin (Taylor, 2003). At the same time, some percents of tritium atoms reversibly replaced H in OH, NH and SH bonds of organic molecules in the body, while about 1% of the activity is incorporated into stable CH bonds. The physical and chemical properties of tritium are almost the same as that of hydrogen.

The biological half times of tritiated water (HTO) are 10 days (97%) and 40 days (3%), whereas for OBT are 10 days (50%) and 40 days (50%) (ICRP, 1997 & Taylor, 2003). A major part (>90%) of an OBT intake is oxidized and excreted as HTO, this tritium is called the exchangeable tritium fraction. The nonexchangeably bound tritium fraction is normally released only as a result of enzymatic breakdown of the molecules containing this carbon-tritium bound. The HTO is excreted in urine, feces, sweat, and breath in which about a half is excreted via urine.

The tritium toxicity in the human body is contributed by $\beta$ particle emitted by tritium bonded in biochemical structure of the cells that leads to genetic mutation and oncogenic effect of the long live cells. Such as it was described previously, the existence of tritium in the environment possibly lead to the internally radiation effect to human body. Therefore, the existence of tritium in the environment should be monitored. The environmental tritium monitoring generally is carried out by taking environmental materials samples, such as air, water, soil, and plants for measurement using liquid scintillation technique.

1.2 Tritium analysis of environmental samples

1.2.1 Sample preparation

The tritium concentration in the water fraction as well as in the organic fraction of the sample is measured in the form of water molecule. Hence the sample preparation is needed in order to isolate the tritium from the samples bulk and convert to liquid form to be possible for LSC measurement. Distillation is one suitable technique to be applied in that purpose.

The sample is heated and the vapor is condensed to obtain the liquid form of free water tritium. In the case of OBT the sample needs to be combusted and the vapor released is condensed at low temperature (Tjahaja et al., 2004). Some modifications in distillation technique have been developed to facilitate the tritium samples preparation such as described in the following sections.
1.2.2 Sample measurement for tritium

The tritium in the environmental and human body samples are measured using liquid scintillation technique after it is prepared according to its chemical form. In this technique the tritium in the water molecule form is mixed with a liquid detector called scintillator to detect the emitted $\beta$ particle, and then measured using Liquid Scintillation Counter device. In this measurement, $\beta$ particle from tritium interact with the molecule of the liquid detector and produce photon emission which then is detected by a photo multiplier tube in the device. In order to obtain good measurement result it is important to prepare the environmental sample in the form of clear and colorless liquid (Rusconi et al., 2006; Hariharan & Mishra, 2000). Therefore, the sample distillation is an important step in the environmental tritium analysis.

Two mL of liquid from prepared environmental sample is added to 13 mL scintillator (as liquid detector) in a 20 mL scintillation vial. The mixture is shaked vigorously and kept in the dark and cool storage for 24 hours to obtain the stable mixture. The tritium content in the environmental samples is measured using LSC for 1 hour.

2. Separation of tritium from the environmental samples

2.1 Separation of tritium from air

The measurement of tritium in the atmosphere is begun with air sampling where the solid absorbent such as silica gel and molecular sieve generally used to collect air sample. The silica gel is an oxide silicate having bee nest like structure, porous, and has very wide surface (Patton et al, 1995). Each gram of silica gel has great affinity toward water molecule, so it is very effective for water absorption. Generally, the silica gel contains blue cobalt salt as an indicator of water content that can exhibit color change from blue to pink after absorbing water or water vapor. The disadvantage of silica gel use in atmospheric tritium sampling is the bond of tritiated water to silica gel grain that is take place in the surface of the grain causing an easy evaporation if it is used in high temperature environment (Patton et al, 1995).

The other solid absorbent can be used for atmospheric tritium sampling is molecular sieve that made of aluminosilicate (zeolite). The aluminosilicate is a chelat compound that can absorb the chemical substances both organic and inorganic as well as water molecule to the cavity of its molecular structure (Iida et al., 1995). The cavity of molecular sieeve structure have various sizes such as 3Å (MS3A), 4Å (MS4A), 5Å (MS5A) and 10Å (MS13X).

The sampling apparatus for tritiated water vapor collection is composed of a sucking pump, a quartz tube of 0.80 m length and 0.05 m diameter, and a water bubler bottle installed at the end of the circuit to confirm the air flow (Figure 1). The atmospheric air is drawn by the pump with low flow rate of about 15 L/h and flown to the tube packed with the silica gel or molecular sieve pelet. Sampling is conducted for about 15 h, the time when the 50 g packed absorbent has been saturated absorbing the tritiated water vapor.

The tritium water vapor absorbed in the solid absorbent is then recovered by developed distillation technique using an apparatus shown in Figure 2. The distillation apparatus is composed of nitrogen gas source, a tube furnace, and a condensation system. The condensation system consists of two condensation tubes that placed in a serial arrangement.
Fig. 1. The apparatus for tritiated water vapor sampling in an ice box. The quartz tube with the tritiated water vapor saturated absorbent is inserted into the tube furnace, and connected to nitrogen gas source using teflon pipe, while the other side is connected to the condensation system. The solid absorbent is heated at 400 °C under nitrogen gas flow until stable mass of solid absorbent is gained. For 50 g of absorbent about 24 h heating is needed. The vapor released from the absorbent is flowed by nitrogen gas to the condensation tubes in the ice box and condensed to liquid form that trapped in the bottom of the tubes. The trapped water is then ready to be measured using liquid scintillation counting method, such as described in section 1.2.2.

The whole system, both tritiated water vapor sampling and separation method have been tested by introducing the various activities of tritiated water vapor to the air sampling apparatus for 15 h. The absorbed tritium in the solid absorbent is then recovered using the distillation apparatus in Figure 2. The measurement of the recovered tritiated water indicates the linearity correlation between the tritiated water vapor radioactivities with those in the recovered water.

Fig. 2. Distillation apparatus to separate tritiated water vapor from the absorbent bulk.
Figure 3 and 4 show the linear relationship between the tritiated water radioactivity in the air with those recovered from the absorbent with the correlation coefficient of 0.985 and approximately 1 for silica gel and molecular sieve, respectively. The capacity of silica gel and molecular sieve in tritiated water vapor absorption has been tested gravimetrically, and the capacity of 48.6% and 43.6% were obtained for silica gel and molecular sieve, respectively.

![Graph](Fig. 3. The linear relationship between the tritiated water activity in the air with those recovered from silica gel)

![Graph](Fig. 4. The linear relationship between the tritiated water activity in the air with those recovered from molecular sieve.)

2.2 Separation of tritium from water

As stated in previous section, the tritium measurement using LSC method needs clear and colorless liquid, hence the environmental water sample with low organic matter can
directly prepared using simple water distillation. The distillation should be conducted in relatively low temperature of about 70 °C and the water have to be completely distilled to near dryness. The complete distillation is necessary as tritium which has relatively higher atomic mass compared to its isotopes (hydrogen and deuterium) will be distributed in the bottom of the distillation flask and evaporated later. For the environmental water samples with high organic matter content, the distillation should be done with the addition of KMnO₄ and activated carbon to decompose and absorb the organic impurities. The organic matter content in the water samples can be determined visually or using UV vis spectrophotometery method. The complete distillation is carried out at 70°C temperature.

The apparatus for environmental water sample distillation is shown in Figure 5. The apparatus is composed of round bottom flask for water samples, a glass water circulation condensor, a glass vessel with a valve in its end opening for collecting distillate, an adapter between the sample flask and the condensor that equipped with a thermometer. In the top end of the condensor vessel a glass vessel filled with silica gel absorbent is installed to avoid the laboratory atmospheric water vapour contaminates the distillate obtained. The sample heating is conducted using a mantle heater.

In order to obtain the clear and colourless distillate, the distillation can be done once, but if the water samples contain high organic matter the distillation is conducted more than once until the clear and colourless distillate is gained.

![Distillation Apparatus Diagram](https://www.intechopen.com)

Fig. 5. The distillation apparatus for environmental water samples preparation
2.3 Separation of tritium from soil

Tritium in the soil is present as both free water tritium (FWT) and organically bound tritium (OBT). The soil tritium is separated from the soil bulk using modified distillation technique to obtain tritium samples as water to be possible for LSC measurement.

2.3.1 Free water tritium

The FWT sample from the soil represents tritium in the water molecule in soil particle surface and interstitial particles (Tjahaja et al., 1995). The FWT from soil sample is collected using distillation technique by heating the soil sample at 150 °C in a quartz tube and condensing the vapor released from the samples at low temperature of about 10 °C. The sample heating is conducted in a tube furnace. The distillation apparatus for soil FWT collection consists of oxygen and nitrogen gas flow system, tube furnace, and condensation system as shown in Figure 6. The condensation system consists of two condensation tubes in serial arrangement inside an ice box. A quartz tube packed with 250 g of soil samples is inserted into the furnace and connected to the gas source system in one side and to the condensation tube in another side. The soil sample is heated at 150 °C under the slow nitrogen gas flow and the vapor from the heated soil is transferred to the condensation tube to be condensed in liquid form. The distillation process of 250 g soil sample take about 30 h, nevertheless it depends on the soil water content. The FWT obtained from this process possibly contains organic material that is visually seen as unclear and yellow color liquid. In this condition, FWT distillation is necessary to remove the impurities of the FWT so clear and colorless water can be obtained.

2.3.2 Organically bound tritium

The dried soil sample as the residue of the FWT distillation process is combusted in the same quartz tube by increasing the temperature of the furnace to 450 °C and applying the oxygen gas to the tube to obtain OBT as liquid form. The combustion results (H₂O and CO₂) is flowed to the condensation tubes where the H₂O gas will be condensed as liquid while the
CO₂ will be flushed out since its condensation point is lower than the temperature in the condensation system. The combustion is held for 30 hours until all soil samples become ash. The condensed water (OBT) contains the organic matter, especially if the combustion process is not perfect. Therefore, a distillation with KMnO₄ and activated charcoal addition is necessary to obtain the clear and colorless water for the LSC measurement.

The performance of the distillation apparatus for soil tritium separation has been tested by preparing 250 g of soil samples with varied tritium concentration, i.e. 180, 270, 555, and 740 Bq. A good performance of this apparatus system is indicated by the recovery efficiency of 92% for total tritium (FWT and OBT) in the soil. The measured tritium activity in soil samples both for FWT and OBT linearly increased with the amount of given tritium activity in the soil. The correlation of tritium activities in the soil with those measured after sample preparation shows linear correlation with the coefficient of 0.99, such as shown in Figure 7.

The distillation of FWT and OBT after the heating and combustion process is necessary to be conducted to obtain the suitable sample for LSC measurement. The distillation of FWT and OBT water after the sample heating or combustion can increase the tritium analysis efficiency of about 10%.

Fig. 7. The correlation of tritium radioactivity in the soil with those measured (FWT and OBT) after sample preparation.

2.4 Separation of tritium from plant

As the same as with the soil, the tritium radionuclide in plant occur in the form of free water tritium and organically bound tritium (Konig, 1990), hence the sample preparation for tritium measurement using LSC method is the same as with those for soil sample.

The performance of the apparatus for tritium separation from plant has been tested (Tjahaja et al., 2005) using varied tritium concentration in the grass of 225, 450, 670, and 960 Bq in 230 g of fresh grass. The tritium contaminated fresh grass was packed in a quartz tube and heated at 150 °C for 30 h using tube furnace that was arranged as in Figure 6. The vapor released was condensed in condensation tube cooled by ice cube. The recovered vapor as FWT was completely distilled by KMnO₄ and activated charcoal addition until clear and
colorless distillate was obtained and could be measured using LSC technique. The dried grass sample in the quartz tube was then combusted at 450 °C under oxygen gas flow using tube furnace in the same apparatus. The gas released (H₂O and CO₂) from the combusted grass samples was flowed to the combustion tube and condensed at ice temperature. The condensed water representing the OBT of the grass as the same as the FWT was completely distilled and measured using LSC method.

The increase of tritium activity in the grass sample is followed with the increase of the recovered FWT and OBT. The correlation of tritium radioactivity in the grass with those measured after sample preparation shows linear correlation with the coefficient of 0.98, such as shown in Figure 8. A good performance of this apparatus system is indicated with the recovery efficiency of 80% for total tritium in the grass (FWT and OBT). The performance of the separation method for plant samples relatively low compared with those for soil samples relating to the complexity of organic compound in the grass leading to the incomplete combustion process in OBT recovery.

2.5 Separation of tritium from human urine

The presence of tritium in the air, drinking water, foods, and other consumer products is the source of human contamination with tritium (Puhakainen et al., 2007 and Trivedi et al., 2000). The detection of tritium contamination in the human body can be carried out using urine samples measurement. Nevertheless, tritium measurement in urine has a limitation relating to the body metabolite content in the urine that significantly affect the LSC measurement (Trivedi et al., 2000; Rusconi et al., 2006). The measurement of such urine possibly to increase counting rate of tritium in the LSC measurement because of chemical luminescence from samples impurities and from β radiation emitted by ¹⁴C bounded in organic matter of body metabolite (Rusconi et al., 2006; Groning, 2004). Such a counting result cause over estimation of tritium concentration in the urine especially in low level tritium measurement (Groning, 2004; Momoshima et al., 1986). Therefore, urine sample
preparation is important to be applied before LSC measurement to reduce body metabolite content affecting the measurement result.

Several urine sample preparation methods have been performance tested for low level tritium (Tjahaja, 2009). Six methods, named A, B, C, D, E, and F, were applied to two urine samples already contaminated with tritium in the concentration of 0.5 Bq/mL and 1 Bq/mL. The method efficiency was analyzed based on the tritium activity recovered in the urine sample after the preparation.

In A - E method, sample preparations were applied such as described in the scheme in the Figure 9-13 (Momoshima et al, 1986; Bhatt et al., 2002), while in the F method an amount of urine sample was directly measured using LSC.

![Fig. 9. The scheme of urine sample preparation by A method](www.intechopen.com)
Fig. 10. The scheme of urine sample preparation by B method

Fig. 11. The scheme of urine sample preparation by C method
Fig. 12. The scheme of urine sample preparation by D method

- 3 g KMnO₄ + 0.1 g NaOH
- Complete distillation at 70 °C

Urine distillate

Urine distillate

E method

2 mL urine distillate + 13 mL scintillator, mixed, stored in the dark and cool for 1 day

2 mL urine + 13 mL scintillator

LSC measurement (1 h)

Fig. 13. The scheme of urine sample preparation by E method

Urine distillate (from D method) + 0.25 mL concentrate H₂SO₄ + N₂

Clear and colorless urine

2 mL clear and colorless urine + 13 mL scintillator, mixed, stored in the dark and cool for 1 day

2 mL urine + 13 mL scintillator

LSC measurement (1 h)
The performance test result can be seen in Figure 14 and 15 for tritium concentration in the urine of 0.5 Bq/mL and 1 Bq/mL, respectively. They indicate that the E method has the highest efficiency with the average of 35% and 58% for tritium concentration of 0.5 Bq/mL and 1 Bq/mL. In the F method that is direct LSC measurement, the tritium activity is not detected in the recovered urine as the same as with those for A and B method. The separation procedure shown in Figure 10-13 is carried out using distillation apparatus shown in Figure 5 in section 2.2.

For tritium measurement in the urine the E method is used. The 24 h urine sample is collected in a plastic bottle, and about 25 mL of the sample is added with 1.25 mg of active charcoal, shaked vigorously for about 30 minutes, then filtered using filter paper to obtain clear and colorless filtrate that free of particulate matter. The filtrate is then again added with active charcoal and distilled completely at 70 °C to eliminate the yellow color of the first filtrate that can alter measurement using LSC device (Puhakainen et al., 2007; Momoshima et al, 1986). The distillate obtained is then added with 1.5 g KMnO₄ and 0.05 g NaOH to destruct the urine organic compound, and again distilled completely at 70 °C. The distillate is then added with 0.125 mL concentrate sulfuric acid to destruct carbonate compound resulted from previous reaction. The CO₂ released from the reaction is then flushed out by nitrogen gas flow for about 10 minutes.

Fig. 14. The method efficiency of 0.5 Bq/mL urine sample preparation. Number 1 represents the F method, numbers 2-6 represent the A – E method.
3. Conclusion

As tritium naturally is present in environmental material, human is normally exposed internally with tritium via foods, drinking water, inhaled air, and also skin absorption. The tritium in the human body contributes radiation doses that affecting human health, hence the monitoring of tritium concentration in the environmental samples becomes interesting to avoid tritium intake by human.

This chapter has described the environmental sample preparation needed in tritium monitoring. Several developed distillation techniques in the preparation of environmental sample including air, water, soil, grass and urine have indicated a good performance result. Nevertheless, the sample preparation method for environmental tritium measurement is still interested to be developed to obtain the better performance. It is hoped that the chapter is technically worth in solving environmental and health problems arise in this industrial era.

4. References


Konig, L.A. 1990. Tritium in the food chain, Radiation Protection Dosimetry, 30 2, pp. 77-86.


Distillation modeling and several applications mostly in the food processing field are discussed under three sections in the present book. The provided modeling chapters aimed both the thermodynamic mathematical fundamentals and the simulation of distillation process. The practical experiences and case studies involve mainly the food and beverage industry and odor and aroma extraction. This book could certainly give the interested researchers in distillation field a useful insight.

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