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An innovative distillation device for tritiated water analysis with high decontamination factor

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HIGHLIGHTS

- Provide an easier way to analyze the distillated environmental tritiated water.
- Condense the evaporated water sample by air cooling without using water.
- The tritium loss rate test in this study is lower than that reported in ASTM D4107.
- Excellent decontaminating ability for eliminating potential interference of LSC.

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ABSTRACT

Institute of Nuclear Energy Research (INER) has designed an air-cooling distillation device and got a US patent. The decontamination factor ^{60}Co and ^{137}Cs is above 23,000. Tritium loss rate is one of testing items in ASTM D4107 Standard Test Method for Tritium in Drinking Water. In this study, the 3 levels (high, middle and low level) of tritium concentration of testing samples for the loss rate test were prepared similar to the concentrations reported in ASTM D4107. The loss rate of the high level is −2.37%, the middle is −2.31% and the low level is −2.47%. These results show that the air-cooling distillation device has good performance in the environmental water tritium analysis work.

1. Introduction

Tritium could be produced by cosmic ray interacting with atmospheric gases at a relative constant level (Theodorsson, 1999). At present, the major source of worldwide tritium is nuclear weapons testing that reached the peak in 1963 with a concentration of 150 Bq/l measured in the rain of Vienna (Guétat et al., 2008). The operation of nuclear power plant may raise the tritium level rising significantly in the local area. Boron used in the coolant of PWRs and the control bar of BWRs may be activated by neutron to produce tritium and be released to the environment. CANDUs (CANada Deuterium Uranium reactors) which use heavy water moderator and heavy water coolant have caused higher release of tritium to environment (Kim and Han, 1999). In future, tritium is being considered as fuel of nuclear fusion reactors, such as ITER (International Thermonuclear Experimental Reactor) (Stamoulis et al., 2011).

Tritium which has 1 proton and 2 neutrons in its nucleus with an atomic mass of three is the only radioactive isotope of hydrogen. Tritium also possesses the similar chemical behavior as hydrogen in environment. The major chemical form in the environment is tritiated water (HTO). HTO could be mixed with environmental water and spread through the environment rapidly and is taken by human easily. So, the environmental tritium monitoring will become a more important environmental issue in the future (Amiro, 1997; Davis, 1997; Kim et al., 1997).

Tritium is a pure low energy β emitting radionuclide (E_{max} = 18.6 keV, E_{ave} = 5.7 keV) and is usually determined by liquid scintillation counting. If a water sample was contaminated by tritiated water, it could also be contaminated by other radio-nuclides and will be accompanied with other α, β and β/γ emitting radio-nuclides which would interference with the liquid scintillation counting (LSC). Environmental water samples may contain a lot of chemical and radioactive interferences which may affect radioactivity counting of LSC or increase the background. Therefore the water sample usually is purified by distillation to reduce the interferences of the counting. Institute of Nuclear Energy Research (INER) has designed an air-cooling distillation device and got a US patent. The device is simply consisted of only two

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components, container and radiator without using water for cooling. The setup of the device is easier and faster than traditional distillation device that only need to put the radiator on the container.

The distillation efficiency, loss rate and decontamination factor of the device were tested in this study. The air-cooling distillation device was heated on a hotplate with temperature control. The temperatures of hotplate were set at about 100 °C, 125 °C, 150 °C, 200 °C, 250 °C and 300 °C for testing distillation rate. The collecting time of 10 ml samples distillated at these temperatures are about 140 min, 85 min, 50 min, 35 min, 12 min and 7 min. The temperature for testing decontamination factors of ^{137}Cs and ^{60}Co is set at 200 °C. The concentrations of ^{60}Co and ^{137}Cs were measured by INER's low background HPGe gamma spectrum analysis system. The ^{60}Co concentration of the distilled water sample is below the minimum detectable activity ($MDA = 6.9 \times 10^{-3}$ Bq/ml). The decontamination factor ^{60}Co is supposed to be higher than 23,000 by comparing the MDA and the original concentration (1.6×10^{2} Bq/ml). The decontamination factor of ^{137}Cs is about 27,000. Tritium loss rate is one of testing items in ASTM D4107 Standard Test Method for Tritium in Drinking Water. In this study, the 3 levels (high, middle and low level) of tritium concentration of testing samples for the loss rate test were prepared similar to the concentrations reported in ASTM D4107. The loss rate is the lost fraction of tritium after distillation procedure. The loss rate of high level is −2.37%, the middle is −2.31% and the low level is −2.47% in this study. These results show that the air-cooling distillation device has good performance in the environmental water tritium analysis work with high decontamination factor.

2. Experimental

2.1. The air cooling distillation device

The air cooling distillation device has two major components as shown in Fig. 1. The upper one, named radiator, is made of alloy for cooling and condensing the vaporized water sample with special design of air cooling. The lower component, named container, is modified from glass beakers for vaporizing water sample. A sample collection tube is added to the beaker for collecting condensed water sample.

Mg/Al alloy was selected to make the radiator because that Mg/Al alloy has excellent thermal conductivity and is easier to be processed by CNC machining. The structure design of the radiator is illustrated in Fig. 2. The middle of the radiator bottom is shaped as a protrusion for gathering up the condensed water sample on the radiator bottom and dropping into the sample collection tube of the container. The heat sink blades of the radiator were designed to be an S shape to enlarge the area of the blades and enhance the efficiency of the radiator. All of blades are linked a small cylindrical cavity which is named condensation indicator. A small amount of water would be put into the cylindrical cavity as an indicator for showing the radiator is in a good condition of condensing the evaporated water sample.

2.2. The distillation rate test of different heating temperatures

The consuming time of distillation is considered essentially in the management of laboratory. The sample amount for the tritium measurement of liquid scintillation counting is 10 ml in the routine environment monitoring work of INER. So, the distillation rates at different temperatures were measured according the time needed to get 10 ml distilled sample measured by volumetric cylinder. The heating temperatures of hot plate were set at 100 °C, 125 °C, 150 °C, 200 °C, 250 °C and 300 °C. The result is shown in Fig. 3.

2.3. Liquid scintillation counting

A Packard 2260XL liquid scintillation counter was used in this study. The selected scintillation cocktail is Parkard Ultima Gold LLT. 10 ml of scintillation cocktail is used to mix with 10 ml of water sample by shaking the polyethylene counting vial. The counting time of the Packard 2260XL liquid scintillation counter was set at 100 min. The counting window was set a range between 0 keV and 4.6 keV. The counting efficiency of sample is determined automatically by quenching level (tSIE value) of sample measured by Packard 2260XL liquid scintillation counter. The tSIE value of distilled water sample is usually around 300 and the counting efficiency is about 15% in this study.

2.4. The loss test

A known activity of tritiated water (NIST 4394H) was diluted to three different levels of tritiated water, 19.0785 Bq/ml, 1.6827 Bq/ml, and 0.1680 Bq/ml compared to the activity concentration of 11.39 Bq/ml, 1.01 Bq/ml, 0.26 Bq/ml mentioned in ASTM D4107 Standard (ASTM D4107-91, 1991). These different levels of tritiated water were tested for the loss of distillation by using the distillation device. The test of each level was made three times. Distilled tritiated water sample and its original tritiated water were counted by liquid scintillation counter at the same time. The loss ratio of distilling the tritiated water is determined by comparing the measured activities of distilled solution and its original solution.

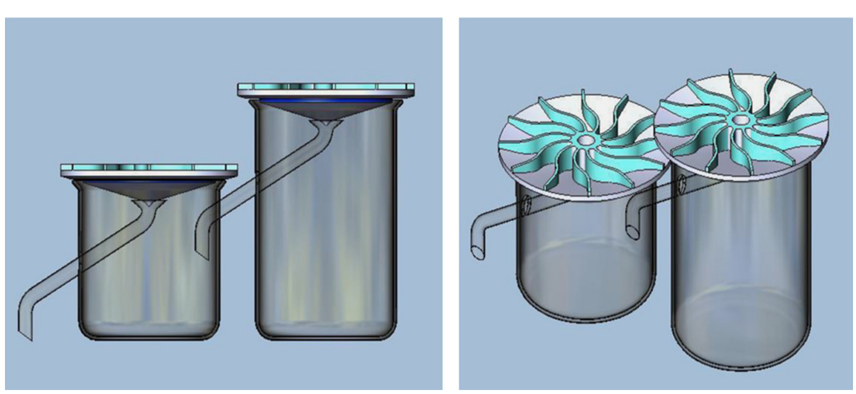

Fig. 1. The air cooling distillation device.