Journal of Environmental Radioactivity 150 (2015) 62–67

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Improving distillation method and device of tritiated water analysis for ultra high decontamination efficiency

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ARTICLE INFO

Article history:
Received 28 January 2015
Accepted 13 March 2015
Available online 22 August 2015

Keywords:
Tritiated water
Distillation
Decontamination efficiency
Gross beta
Air cooling

ABSTRACT

It is important that monitoring environmental tritiated water for understanding the contamination dispersion of the nuclear facilities. Tritium is a pure beta radionuclide which is usually measured by Liquid Scintillation Counting (LSC). The average energy of tritum beta is only 5.658 keV that makes the LSC counting of tritium easily be interfered by the beta emitted by other radionuclides. Environmental tritiated water samples usually need to be decontaminated by distillation for reducing the interference. After Fukushima Nucleaer Accident, the highest gross beta concentration of groundwater samples obtained around Fukushima Daiichi Nuclear Power Station is over 1,000,000 Bq/l. There is a need for a distillation with ultra-high decontamination efficiency for environmental tritiated water analysis. This study is intended to improve the heating temperature control for better sub-boiling distillation control and modify the height of the container of the air cooling distillation device for better fractional distillation effect. The DF of Cs-137 of the distillation may reach 450,000 which is far better than the prior study. The average loss rate of the improved method and device is about 2.6% which is better than the bias value listed in the ASTM D4107-08. It is proven that the modified air cooling distillation device can provide an easy-handling, water-saving, low cost and effective way of purifying water samples for higher beta radionuclides contaminated water samples which need ultra-high decontamination treatment.

1. Introduction

Tritium, the lightest radionuclide, is the known heavest isotope and the only radionuclide of Hydrogen with atomic mass 3. It is one of natural radionuclides produced by the interactions of atmospheric atom with cosmic ray in the upper atmosphere through interactions between nitrogen and cosmic ray (HPS, 2011; Okada and Momoshimat, 1993). A very minor fraction of natural tritium may be produced in the earth's crust by the nuclear reaction of ^{6}Li and neutron from the spontaneous fission of uranium (Kaufman and Libby, 1954). UNSCEAR (2000) estimated that cosmogenic tritium is produced 7.2 × 10^{16} Bq every year and the equilibrium global atmospheric inventory is near 1.275 × 10^{18} Bq. The natural (pre-nuclear age) tritium activity concentrations of rain water were in the range 0.18–1.11 Bq/l (Gateriu and Melintescu, 2010; ANL, 2005). Since 1945, human began nuclear bomb test in atmosphere that made the activity of tritium released to atmosphere exceed the cosmogenic tritium activity. The range of tritium activity released to atmosphere up to 1976 was estimated from 1.2 × 10^{20} Bq to 2.4 × 10^{20} Bq (Bowen and Roether, 1973; Schell et al., 1974; Mitchel, 1976; UNSCEAR, 1977). UNCEAR (1977) adopted the value of 1.7 × 10^{20} Bq (4500 MCi) as the representative amount of the gloabally dispersed tritium from nucleaer bomb test. The atmospheric tritium may be deposited with rain and raise the tritium concentration in surface water and then goes into hydro cycle. The monitoring result of tritium precipitation at Ottawa station reaches the peak value near 1000 Bq/l in 1963. Although the tritium concentration in atmosphere is decreased with time, the tritium contributed from nuclear test is still about 1.4 × 10^{19} Bq, 10 times larger than the natural contribution, remains in atmosphere estimated in 2007 (Gateriu and Melintescu, 2010). Nuclear facilities, including nuclear power plants, nuclear fuel reprocessing plants and tritium production plants, have also contributed tritium to environment that may cause tritium concentration of environmental samples raised in the local area (Janovicsa et al., 2014; de Carvalho Gomes et al., 2014; Kim, 2013; Nankar et al., 2012; Kim et al., 2012; Hughes et al., 2011; Akata et al., 2011; Twining et al.,

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http://dx.doi.org/10.1016/j.jenvrad.2015.03.026
0265-931X/

2011; CNSC, 2009; Bolsunovsky and Bondareva, 2003; Okada and Momoshimat, 1993). HTO measurement is included in the monitoring items for understanding the contamination dispersion of the nuclear facilities.

Tritiated hydrogen (HT), tritiated water (HTO) and organically bound tritium (OBT) are three major chemical forms of tritium exist in the environment. Tritiated water is the most abundant chemical form of tritium in the environment. In natural environment, HT is only about 0.02~0.2% tritium contribution in troposphere and 0.004~0.007% tritium contribution in stratosphere. The tritium contribution of HTO is almost 100% and 90% in hydrosphere (Begemann, 1963; CNSC, 2009). The tritiated hydrogen may be oxidized and become tritiated water through oxidation in air, photochemical oxidation in air, exchange reaction with water, oxidation with soil and oxidation in plants. The material contents in air and the water content in soil may increase the oxidation rate. However, a lot of studies show that the relative rates involving plant systems are far greater than air systems and soil systems (Okada and Momoshimat, 1993). After taken by plant, HTO may be partly released into atmosphere and partly kept by plants as tissue water tritium (TWT) and the partial of the tritium become OBT (Amano et al., 1995; Galeriu et al., 2013). A useful definition, translocation index (TLI), has been defined for studying the translocation of OBT to the edible plant part. TLI is the percentage of the OBT concentration in grain at harvest related to the TWT concentration in leaves at the end of exposure to HTO (Diabaté and Strack, 1997) Most of the published TLI data are far below 1% (Galeriu et al., 2013) that means most of the tritium in the plants exists in the chemical form of HTO. Therefore, monitoring HTO in different environmental samples may investigate the situation of tritium dispersion in the environment.

Tritium is a pure beta radionuclide. The maximum beta particle energy of tritium is 18.6 keV (NCRP, 1985) which is very low compared to the energy of beta particle emitted from other radionuclides. The radioactivity measurement of HTO usually uses liquid scintillation counting (LSC) because the beta particles may release their energy directly in the scintillator and produce the counting signal that make the counting efficiency higher than other methods. Environmental samples may contain a lot of beta radionuclides which may increase the counting background of LSC as well as some chemicals which may interfernce the counting efficiency of LSC (Theodorsson, 1999). Therefore, environmental water samples usually need to be decontaminated by distillation for reducing these interferences. The decontamination efficiency may be needed to be higher than traditional distillation device. For example, a lot of the gross beta measurement results of groundwater obtained around Fukushima Daiichi Nuclear Power Station after Fukushima Nuclear Accident are over 30,000 Bq/l and the highest is over 1,000,000 Bq/l (TEPCO, 2014) that not only can increase the counting background of LSC for tritiated water analysis but also increase the possibility of cross contamination of environmental device for environmental HTO measurement.

This study is trying to establish a sub-boiling distillation method and improve the distillation device for improving decontamination efficiency that may overcome the interference of high gross beta concentration in environmental water likes the groundwater samples obtained around Fukushima Daiichi Nuclear Power Station. Institute of Nuclear Energy Research (INER) had made an air cooling distillation device which got a United States patent (US 7,927,462 B2) in 2011. The device has a flat bottom water sample container which can be heated by hot plate to replace the round bottom flask used in conventional distillation. The vapor from the heated water sample can be condensed on the radiator covered on the top of the container. The radiator has a protrusion which can help the condensed water drop into the inlet of the conduit penetrating the container for the distilled sample collection. The DFs of the distillation device had been tested for Cs-137 and Co-60. Both of the DFs are above 20,000 for Cs-137 (Fang et al., 2013). This study tried to increase the height of the container and modify the heating process of distillation which would be well controlled under a sub-boiling condition. The temperature range of heated water samples for distillation was kept from 80 °C to 90 °C measured by infrared thermometer in this study. The sub-boiling distillation rate of this method is about 10 ml/h to 15 ml/h that may be influenced by environmental condition. The DF value of Cs-137 is improved from about 20,000 to about 450,000, above 20 times.

The vapor pressure isotope effect (VPIE) of HTO is needed to be considered that may make the HTO concentration of distilled water sample decreased in the distillation procedure (ASTM, 2013; Atkinson et al., 2014). This study also designed a set of experiments for understanding the VPIE of HTO in the distillation procedure and evaluating the bias correction factor for the tritium analysis of water sample by using the air cooling distillation device. The result could be better than the listed value in ASTM D4107-08 (ASTM, 2013).

2. Methods and experiments

2.1. The air cooling distillation device and the modification of the container

The air cooling distillation device has two major components: container and radiator (Fig. 1). Water sample can be put in the container which looks like a glass tube with a flat bottom. There is a conduit penetrating the container. The conduit has an inlet located at the center of the container's upper part water sample and an outlet outside the container for collecting condensed. The radiator has a lot of heat sink blades for dispersing the heat from condensing the vaporized water sample. The radiator is made of Mg/al alloy because Mg/Al alloy has excellent thermal conductivity and is easier to be processed by CNC machining than other kinds of metal and alloy. There is a cylindrical cavity on the center of the heat sink blades. The cylindrical cavity can be used as a simple overheating indication if it was filled with water. The heat sink blades were designed in an S curve shape to enlarge its heat sink area. The radiator has a taper bottom with a protrusion furnished at center as a cusp. The condensed water may flow to the cusp and drop into the

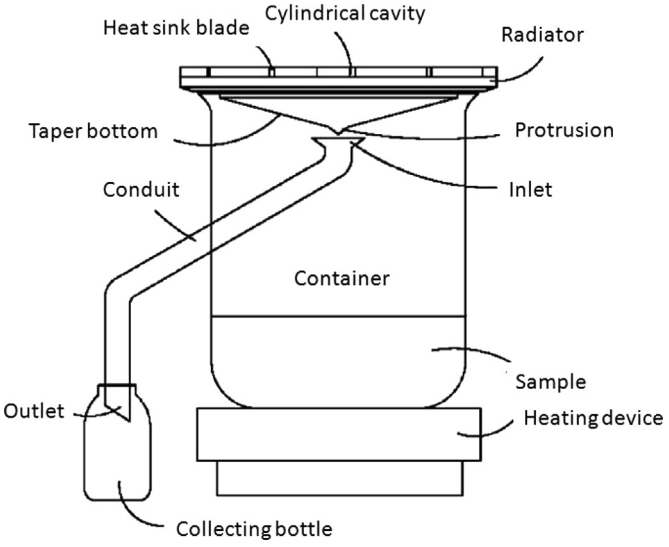

Fig. 1. The components of the air cooling distillation device.