Determination of Organically Bound Tritium (OBT) in a Wheat Sample and Its Interlaboratory Comparison Study

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1. Introduction

Tritium (^{3}H) is released from nuclear power plants during their normal operation or decommissioning, and may cause mutation of the cell and/or death of the cell in human tissues or organs by its inhalation and ingestion [1]. Organically bound tritium (OBT) has become of increased interest within the last decade, with a focus on its behavior and also its analysis, which are important to assess tritium distribution in the environment [2]. OBT is formed in living systems through natural or biological processes from HTO [3]. OBT is sum of the exchangeable (E-OBT) and non-exchangeable forms (NE-OBT) [4]. In this study, the separation of ^{3}H in the OBT sample was carried out by using a combustion/absorption method [5]. To determine radioactivity of ^{3}H in the OBT sample, produced HTO during the combustion of the sample was trapped in a dilute HNO_3 solution. The HTO trapped solution was mixed with LSC cocktail to determine the radioactivity concentration of ^{3}H in the sample by using a liquid scintillation counter. In addition, the obtained radioactivity concentration of ^{3}H and our method were compared with those of other international laboratories.

2. Materials and Methods

2.1 Equipment and Chemicals

The combustion system (Pyrolyser-6 Trio^{TM}) was purchased from RADDEC. 0.1 M HNO_3 was prepared by dilution of concentric HNO_3 (Merck) with deionized water (DI water, 18.2 MΩ·cm, Milli-Q Direct 8 water system). Gold Star Multi-Purpose Liquid Scintillation Cocktail (Meridian) was used as a scintillation cocktail. 0.5% Pt on alumina pellet, sea sand (30 ~ 50 mesh), and quartz wool were bought from Elemental Microanalysis, Samchun Pure Chemical, and Grace Davison Discovery Science, respectively.

2.2 Combustion of Wheat Sample

The combustion equipment and analysis for a wheat sample is depicted in Fig. 1. The dehydrated wheat sample was supplied by CEA (French Alternative Energies and Atomic Energy Commission, France). The sample boat including ca. 5 g of wheat sample was placed in a silica glass work tube within the sample oxidation zone of the furnace, and heated using a preset temperature profile (Fig. 2) with air purge. After the temperature of the sample oxidation zone has reached higher than 500 ºC, oxygen was purged into the combustion furnace system to burn the incompletely combusted sample. The formed gases pass through the 0.5% Pt/alumina catalyst pellets packed column to enhance the complete oxidation of the gases to H_2O and THO. H_2O and THO are trapped in a bubbler containing 20 mL of 0.1 M HNO_3.
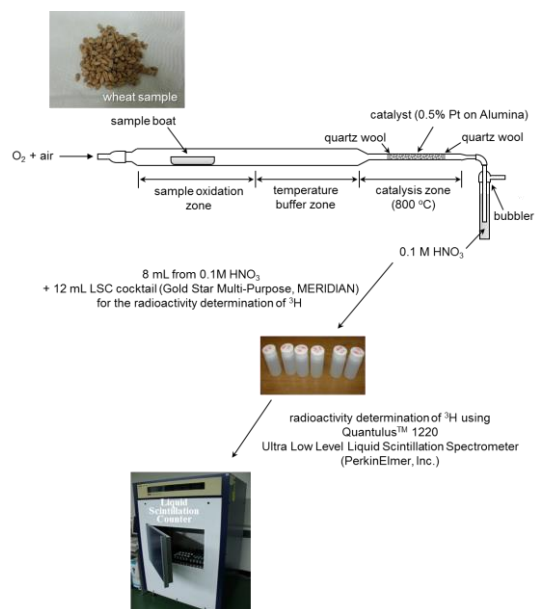

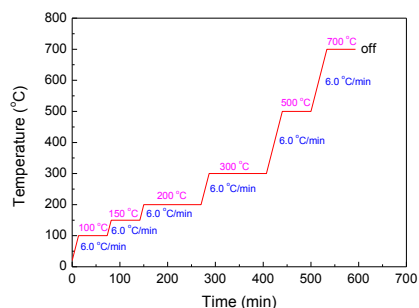Fig. 1. Schematic diagram of combustion/absorption and analysis system.

Fig. 2. Temperature profile of the sample oxidation zone.

2.3 Determination of ^{3}H in Sample

Ca. 8 g of H_2O (including THO) vapor trapped 0.1 M HNO_3 solution in the bubbler was transferred to a 22

mL plastic LSC vial, and mixed with 12 mL of Gold Star Multi-Purpose Liquid Scintillation Cocktail. Radioactivity of ^{3}H in the vial was determined by a Quantulus^{TM} 1220 Ultra Low Level Liquid Scintillation Spectrometer (PerkinElmer). The radioactivity (A_s) of ^{3}H in the sample was calculated by the following equation:

$$A_s = \frac{(C_s - C_B)}{60} \frac{100}{\varepsilon} \frac{1}{m_s} \frac{(m_f - m_t)}{m_b} \frac{100}{R} \quad (1)$$

where A_s, C_s, C_B, ε, m_b, m_f, m_t, m_s and R are the radioactivity concentration in the sample, the sample count rate, the background count rate, the counting efficiency for LSC, the mass of the bubbler solution taken for analysis, the final bubbler mass, the bubbler tare mass, the mass of sample and the furnace recovery, respectively.

The counting efficiency for LSC was measured using an external standard method (SQP(E)) at different quench level as represented in Fig. 3.
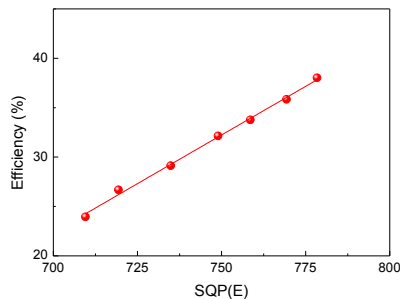

Fig. 3. Counting efficiency curve of ^{3}H for different quench levels (SQP(E)).

The uncertainty (U_{As}) quoted is the expanded uncertainty with a coverage factor $K = 2$ (95% confidence level). Uncertainty in ^{3}H concentration is incorporating uncertainties from the counting statics, mass, signal reproducibility, and furnace recovery. The radioactivity concentration and its associated uncertainty with a coverage factor of $K = 2$ is then $A_s \pm 2 \times U_{As}$ Bq/g.

3. Results and Discussion

In the combustion method for the determination of ^{3}H in the sample, the complete oxidation of sample is one of the significant factors to obtain reliable radioactivity of the sample. After the combustion of the wheat sample (OBT sample), any ash was not remained in the sample boat owing to the complete oxidation of the sample. The radioactivity concentration of ^{3}H in the dehydrated wheat sample was obtained with 10 duplicate wheat samples. The obtained average value with the expanded uncertainty (a coverage factor $K = 2$, 95% confidence level) was 37.3 ± 15.0 Bq/kg.

More than 25 laboratories attended for the OBT interlaboratory comparison study from and Canada,

United Kingdom, France, Japan, Romania, Belgium, South Korea, etc. To determine the radioactivity concentration of ^{3}H in the wheat sample, the laboratories used various LSC cocktails (UltimaGold LLT, Hisafe 3, Goldstar and Monophase S), combustion methods (combustion bomb, tube furnace and oxydiser) and measurement equipments (Quantulus 1220, Tricarb, ALOKA LB5 and mass spectrometer). The results obtained by the laboratories were shown in Fig. 4, and their average value was 42 ± 14 Bq/kg. The result was very good although one lab is slightly over the confidence interval.
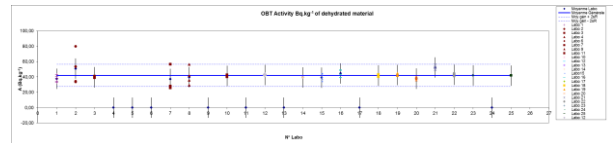

Fig. 4. Graphical presentation of the interlaboratory comparison result.

3. Conclusions

In conclusion, we have determined ^{3}H in the wheat sample by the combustion of the sample, the oxidation of produced gases from the pyrolyzed sample, and LSC analysis of the scintillation cocktail mixed 0.1N HNO$_3$ solution absorbing HTO produced by the oxidation of gases. The combustion method was easy and rapid to analyze OBT in the sample. The minimum detectable activity (MDA) of the combustion method was 16 ~ 18 Bq/kg. Our result (37.3 ± 15.0 Bq/kg) was similar to other international laboratories' results although they used different LSC cocktails, combustion methods and measurement equipments.

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