Development of an active tritium sampler for discriminating chemical forms without the use of combustion gases in a fusion test facility

Masahiro Tanaka[a,b,*], Hiromi Kato[a], Yukie Yamamoto[a], Chie Iwata[a]

[a] National Institute for Fusion Science, Oroshi-cho, Toki, Gifu 509-5292, Japan
[b] SOKENDAI (The Graduate University for Advanced Studies), Oroshi-cho, Toki, Gifu 509-5292, Japan

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ABSTRACT

A new type of active tritium sampler that can discriminate between chemical forms in a fusion test facility without the use of combustion gases was developed. The proposed tritium sampler was operated using water vapour instead of combustion gases. To test the operation and performance of the device when water vapour is used, we evaluated the catalytic oxidation properties, and the evaporation and collection of water vapour under actual sampling conditions. The properties of the added water mass and the operation temperature of catalysts in the proposed sampling system were then determined. Thereafter, we carried out air sampling for tritium monitoring. The levels of tritium concentration measured by the proposed tritium sampling system were similar to the values measured by the conventional sampling system. Our findings show that the proposed tritium sampling system without combustion gases is a good replacement for the conventional tritium sampling system in a fusion test facility.

1. Introduction

Tritium is a radioactive isotope of hydrogen that emits beta rays with a maximum energy of 18.6 keV, decays to [3]He, and has a half-life of 12.3 y (DOE HANDBOOK, 2008). Tritium forms naturally through cosmic ray interactions with the atmosphere. The annual production rate is estimated to be 70 PBq, and the resulting tritium inventory in nature is calculated to be 1–1.3 EBq. Furthermore, tritium is produced in nuclear power reactors during the fission of heavy nuclei and during neutron interactions with coolant materials. The amount of tritium discharged from nuclear facilities from 1995 to 1997 is estimated to be 43 PBq (UNSCEAR, 2000). Further, tritium is also a by-product of atmospheric nuclear tests. Based on the production rate and the total fusion and fission yield of nuclear tests, the production of tritium during nuclear tests is estimated to be 185 EBq. Since the tritium produced during atmospheric nuclear tests decays over time, the present level is 19 EBq (Bennett, 2002). In the1970s, the atmospheric tritium concentration was more than 100 mBq/m^3 as a result of nuclear testing during the 1950s and 1960s, but it has now exponentially decreased to less than 1/10 of its peak.

Nuclear fusion, for which deuterium (D) and tritium (T) are utilized as fuel, is considered a key energy source. These hydrogen isotopes can easily permeate metal walls under elevated temperatures. The leakage of tritium from fuel processing systems in a fusion facility cannot be entirely stopped. For safety against tritium and radiation management, tritium is removed from the air in facility rooms by tritium removal systems. In accordance with radiation management regulations, part of this tritium is released into the environment from a stack. As a monitoring technique, a tritium sampler is used to accumulate the extremely low levels of tritium within a given period. Koarashi et al. (Koarashi et al., 2004, 2006), monitored total tritium levels using an active sampler at a fuel reprocessing plant.

Measuring and monitoring the different forms of tritium is preferred as the biological effects of tritium depend on its chemical form. In tritium samplers that discriminate chemicals, combustion gases such as hydrogen and methane are required to obtain water samples sufficient for liquid scintillation counting because atmospheric concentration levels of H_2 and CH_4 are very low at about 0.53 ppm (Novelli, 1999, Price, 2007) and 1.75 ppm (Dlugokencky, 2003), respectively. In the radiation control area of a fusion test facility, the tritium sampler has to operate without any supervision. Therefore, it is imperative that the system functions safely when combustion gases are used. With these considerations, we have developed a continuous tritium sampler that uses water vapour instead of combustion gases, based on the work of Kakiuchi et al. (Kakiuchi, 2007). We also evaluated the efficiency of water evaporation from a vial, and the oxidation performance of the catalysts under very low concentrations of hydrogen and methane in relation to temperature in this work. In addition, we present the
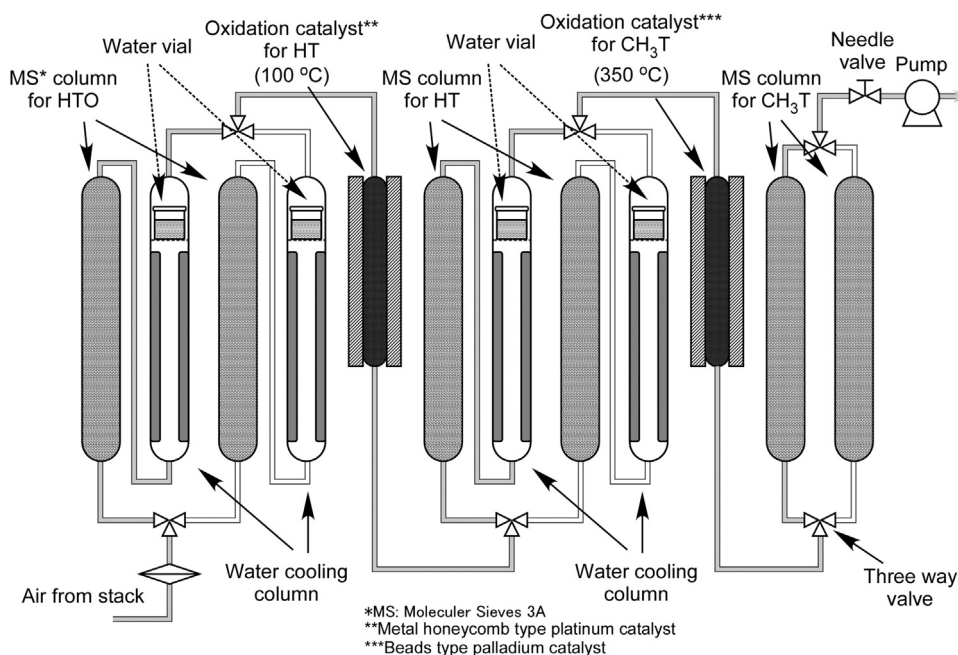

Fig. 1. Schematic diagram of the active tritium sampling system.

monitoring results for actual stack air sampling.

2. Experimental apparatus and method

2.1. Tritium sampling and activity measurement

A tritium sampler system was installed in the machine room of the building in which the fusion test device was housed and it was connected to the sampling line in the stack. The fusion test device has been operated since the last decade to study the plasma physics (Komori, 2010). It has not, however, been used to conduct plasma experiments using hydrogen isotope gases like deuterium. While naturally formed tritium is present, artificial tritium produced by fusion reactions is absent in the fusion test device building.

A schematic diagram and photographs of the developed tritium sampler are shown in Figs. 1 and 2, respectively. The sampler has two paths for continuous sampling; the operational path switches after a given period of time. Air sampling is done at a flow rate of less than 2000 (std) cm³/min for a few weeks. The total amount of air collected was approximately 4–20 m³.

Molecular sieves (Wako Pure Chemical Industries, LTD.) of 3A-type one-sixteenth inch pellet were used. The dry molecular sieve was packed with approximately 550 g of absorbent. Tritium in the chemical form of HTO was collected in the first column of water vapour in the sampled air. After drying the air using molecular sieves, tritium free water vapour was added by evaporating tritium free water to the sampled air. Twenty to forty grams of tritium free water was added to an acryl vial and set in the water cooling column. The temperature of cooling water was controlled and maintained between −10 °C and 10 °C. Tritium in the chemical form of HT was then converted into HTO in an oxidation furnace with metal honeycombs supporting the platinum catalyst (Tanaka Kikinzoku Industry, Pt: 6 g/L, φ25mm and 150 mmL, cell density: 300 CPSI) at 100 °C. The HTO formed was then collected together with tritium free water in the second molecular sieve column. Lastly, the tritium free water vapour was added by evaporating the tritium free water to the sampled air in the same manner as in the case of HT collection. The residual tritium in chemical form of CH₃T was then converted into HTO by an alumina supported palladium catalyst (NE Chemcat, ND-101, Pd: 5 g/L, packed weight: 290 g, column shape: φ40 mm (O.D.) and 300 mmL) heated to about 350 °C.

The HTO was collected together with tritium free water in the third molecular sieve column. The water collected in the molecular sieve columns was desorbed at a temperature of 400 °C and then recovered using a cold trap of about 2 °C under an N_2 gas purge at a rate of 1 L/min for 3.5 h.

To measure the tritium activity, 65 mL of the recovered water from the HTO collection column and 65 mL of liquid scintillator Ultima-Gold LLT (Perkin Elmer Co. Ltd.) were mixed in a 145 mL Teflon vial. Ten millilitres of the water recovered from each of the HT and CH₃T collection columns were mixed with 10 mL of liquid scintillator in 20 mL Teflon vials. Background samples in 145 and 20 mL vials were prepared using tritium free water. After leaving these samples for a few days in a low background liquid scintillation counter LB-7 (Hitachi - Aloka Co. Ltd.), the tritium activity was determined for a total of 1500 min per sample.

2.2. Control of evaporation of water

The cylindrical water vial was set in the water cooling column to control the evaporation temperature, as shown in Fig. 2. The rate of evaporation from the cylindrical water surface was estimated by following the theoretical evaporation equation derived from natural convection condition (Ueda and Shiba, 1964):

$$w_{loss} = 3.2 \times r^{1.75} \times (e_s - e) \times t \times 10^{-3}, \quad r < 10 cm$$
$$\approx 3.2 \times r^{1.75} \times e_s \times t \times 10^{-3} \quad (1)$$

where, w_{loss} [g/h] is the rate of evaporation from the water surface, r [cm] is the radius of the cylindrical water vial, t [h] is operation time, e_s [hPa] is saturated water vapour pressure at a given temperature, and e [hPa] is the atmospheric water vapour pressure. In this case, the value of "e" can be ignored because the sampling gas passing through the molecular sieves column is in an extremely dry condition (less than 0.1 Pa). According to Eq. (1), the amount of evaporation from the cylindrical water vial is proportional to the surface area of the water vial, and sampling time and saturated water vapour pressure act as functions of ambient temperature.