

# Determination of tritium activity and chemical forms in the exhaust gas from a large fusion test device

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## **Title page**

2 Names of the authors: Masahiro Tanaka<sup>1, 2</sup>, Hiromi Kato<sup>1</sup>, Naoyuki Suzuki<sup>1</sup>, Chie Iwata<sup>1</sup>,  
3 Naofumi Akata<sup>1, 2</sup>

4 Title: Determination of tritium activity and chemical forms in the exhaust gas from a large  
5 fusion test device

6 Affiliation(s) and address(es) of the author(s):

7 1. National Institute for Fusion Science, National Institutes of Natural Sciences, Oroshi-  
8 cho, Toki, Gifu 509-5292, Japan

9 2. SOKENDAI (The Graduate University for Advanced Studies), Oroshi-cho, Toki, Gifu  
10 509-5292, Japan

11 E-mail address of the corresponding author: tanaka.masahiro@nifs.ac.jp

12

13 **Determination of tritium activity and chemical forms in**  
14 **the exhaust gas from a large fusion test device**

15 Masahiro Tanaka<sup>1,2</sup>, Hiromi Kato<sup>1</sup>, Naoyuki Suzuki<sup>1</sup>, Chie Iwata<sup>1</sup>, Naofumi Akata<sup>1,2</sup>

16 <sup>1</sup>*National Institute for Fusion Science, National Institutes of Natural Sciences, Oroshi-*  
17 *cho, Toki, Gifu 509-5292, Japan*

18 <sup>2</sup>*SOKENDAI (The Graduate University for Advanced Studies), Oroshi-cho, Toki, Gifu*  
19 *509-5292, Japan*

20 **Abstract**

21 A water bubbler system that can distinguish chemical forms of tritium was proposed for  
22 long-term tritium monitoring of the exhaust gas of a large fusion test device. The  
23 characteristics and performance of the water bubbler system were evaluated under  
24 operational conditions and confirmed to be suitable for tritium monitoring. For the tritium  
25 measurements, the water bubbler system determined the tritium activity and distinguished  
26 the chemical forms of tritium. The tritium activity and chemical forms in the exhaust gas  
27 provided helpful information to understand the tritium behavior in the large fusion test  
28 device.

29 **Keywords**

30 tritium monitoring, water bubbler system, chemical forms, exhaust gas, fusion test device,

31 **Introduction**

32 Nuclear fusion energy research has made steady progress. Fusion test devices use  
33 hydrogen isotope gases, such as deuterium, and during the operation of large fusion test  
34 devices using deuterium gas, tritium is produced in the vacuum vessel by the d(d,p)t fusion

35 reaction. Thus, tritium analysis of the exhaust gas from fusion test devices is important for  
36 understanding the behavior of hydrogen isotopes.

37 In deuterium plasma experiments in the Large Helical Device (LHD) [1-3], a small  
38 amount of tritium is produced in the core plasma. The tritium atoms, the nuclei of which  
39 are called tritons, could be used as a tracer to investigate the tritium behavior. Some tritons  
40 are implanted deeply into the first wall of the vacuum vessel because the maximum triton  
41 energy is 1.01 MeV. Otherwise, the tritons are transported from the core plasma to the edge  
42 plasma region along the magnetic field, and then released in the exhaust as gaseous tritium  
43 with other hydrogen isotopes and working gas via a vacuum pump system. The gas  
44 composition and the chemical forms of the hydrogen isotopes depend on the operation  
45 conditions of the LHD experiments or the plasma parameters. Thus, a synthetic tritium  
46 analysis system is necessary to determine the activity and chemical forms of tritium in  
47 complex gas compositions. Conventional tritium monitoring systems consist of either an  
48 ionization chamber, a proportional chamber, an active or passive tritium sampler using an  
49 adsorbent, or a water bubbler [4-24]. The ionization chamber is often used for tritium  
50 monitoring in tritium handling facilities [4, 5]. However, it is difficult to distinguish other  
51 interfering radionuclide signals, such as radon, from the ionization signal for tritium  
52 monitoring. Thus, the detection limit is higher than that of other monitoring systems.  
53 Proportional counters are linked to the stack or in-line monitor and usually have a lower  
54 tritium detection limit than ionization chambers [4]. However, proportional counters need  
55 proportional gas (Argon with 10% methane counting gas), the running cost of which is  
56 high for long-term operation. As with ionization chambers, changing gas components  
57 affects the operation conditions of the proportional counter. Therefore, conventional  
58 tritium monitoring systems are not always suitable for the exhaust gas from a fusion test  
59 device. For radiation protection and tritium balance studies in fusion test devices, tritium  
60 monitoring that distinguishes tritiated water vapor, tritiated hydrogen gas, and tritiated  
61 hydrocarbons is required, because the regulatory limits for tritium in air differ between the  
62 tritiated compounds [25]. Active tritium samplers that use molecular sieves to distinguish  
63 chemical forms have been developed for tritium monitoring in stacks and the environment  
64 [8-10, 15-17]. In some samplers [9, 16, 17], the post-sampling processing is labor intensive  
65 because the molecular sieves have to be regenerated at more than 623 K for several hours.

66 To reduce the workload, we propose an accumulated tritium sampling system using a series  
67 of water bubblers combined with two catalysts for distinguishing the chemical forms. The  
68 water bubbler system has some of the advantages: the reduction of the post sampling  
69 processing such as the regeneration of absorbent columns, ease of operation, no addition  
70 of water vapor or combustion gas, and so on. In this paper, we evaluate the performance of  
71 the proposed water bubbler system and discuss the tritium monitoring results for the  
72 exhaust gas from LHD.

### 73 **Tritium monitoring apparatus and analytical methods**

#### 74 **Water bubbler system**

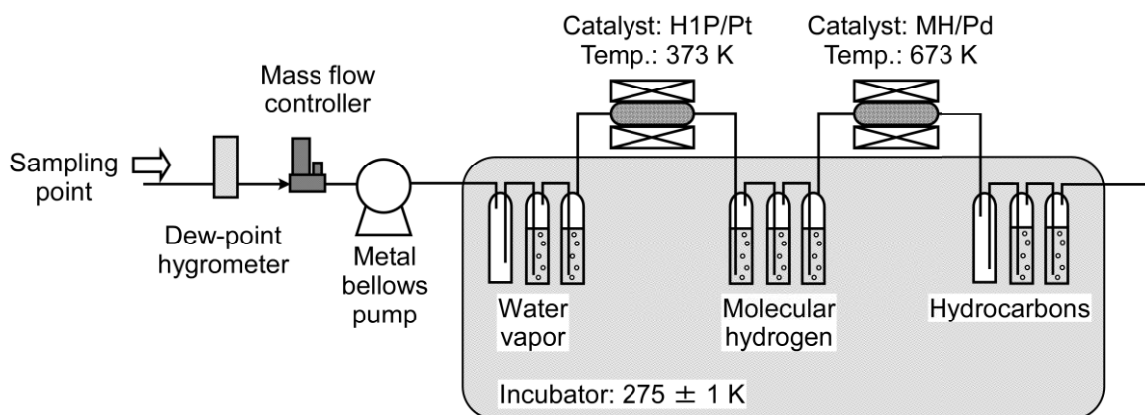
75 A flow diagram of the proposed water bubbler system is shown in Fig. 1. The water  
76 bubbler system consisted of a dew-point hygrometer (Easidew Transmitter, Michell  
77 Instruments Ltd.), a mass flow controller (8500MC, Kofloc), a metal bellows pump (MB-  
78 21, IBS Inc.), a series of water bubbler columns (080100-02, volume: 30 cm<sup>3</sup>, glass filter:  
79 P160, SIBATA SCIENTIFIC TECHNOLOGY LTD.), a two-stage oxidation reactor, and  
80 a low-temperature incubator (MIR-153, SANYO). Each bubbler contained about 19 g of  
81 deionized water produced by an ultrapure water system (Direct-Q UV, Merck Millipore,  
82 Resistivity at 298 K:  $\geq 18.2$  M $\Omega$ -cm, TOC:  $\leq 5$  ppb). The water bubbler column was  
83 installed in the incubator to decrease water evaporation in the bubbler for long sampling  
84 times of more than 1 day. The operating temperature in the incubator was kept at  $275 \pm 1$   
85 K. The gas sampling was performed at a flow rate of less than 200 cm<sup>3</sup>/min for 1 week.  
86 The total amount of air sampled was less than 2 m<sup>3</sup>.

87 Tritium in the chemical form of water vapor was collected in the first bubbler series  
88 when sample gas containing tritium was passed through water. Then, the sample gas was  
89 passed through the low-temperature oxidation reactor to convert the tritiated hydrogen gas  
90 into tritiated water vapor. The low-temperature oxidation reactor was packed with a H1P  
91 hydrophobic platinum catalyst (Tanaka Kikinzoku Industry, Pt: 4 mg/m<sup>3</sup>, packed weight:  
92 290 g, outside diameter [O.D.]:  $\phi 40$  mm, and length: 300 mm) [26]. The hydrophobic  
93 catalyst was chosen to prevent the memory effect and the degradation of the hydrogen

94 oxidation performance under wet conditions at low temperature. The operation temperature  
95 of the oxidation reactor was kept around 373 K. The tritiated water vapor converted from  
96 tritiated hydrogen gas was collected in a second series of water bubbler columns. The  
97 residual tritium in the chemical form of tritiated hydrocarbons was then converted into  
98 tritiated water vapor by a palladium catalyst supported on a metal honeycomb (Tanaka  
99 Kikinzoku Industry, Pd: 4 mg/cm<sup>3</sup>, O.D.: φ26 mm, length: 50 mm, cell density: 300 CPSI)  
100 [27]. The catalyst was heated to about 673 K. Finally, the tritiated water vapor converted  
101 from tritiated hydrocarbons was collected in the third series of water bubbler columns.

102 To determine the tritium activity, water (10 cm<sup>3</sup>) from each of the bubblers was mixed  
103 with liquid scintillator (10 cm<sup>3</sup>, Ultima-Gold LLT, Perkin Elmer Co., Ltd.) in 20 cm<sup>3</sup>  
104 polyethylene vials. The background sample was prepared using deionized water in a 20  
105 cm<sup>3</sup> vial. The deionized water for the background sample was same as ultrapure water used  
106 in the bubbler column. After leaving these samples for several hours in a liquid scintillation  
107 counter (Tri-Carb 4910TR, Perkin Elmer Co. Ltd.), the tritium activity was determined for  
108 a total counting time of 50 min per sample. Counting efficiencies were determined by use  
109 of tritium standard solution (SRM 4361C, NIST, USA). The detection limit of the sample  
110 water was approximately 0.01 Bq/cm<sup>3</sup>. Although the detection limit of the gaseous tritium  
111 concentration depended on the sampling volume, it was less than 10<sup>-6</sup> Bq/cm<sup>3</sup>.

112



113

114 **Fig. 1** Flow diagram of the water bubbler system for distinguishing chemical forms of  
115 tritium.

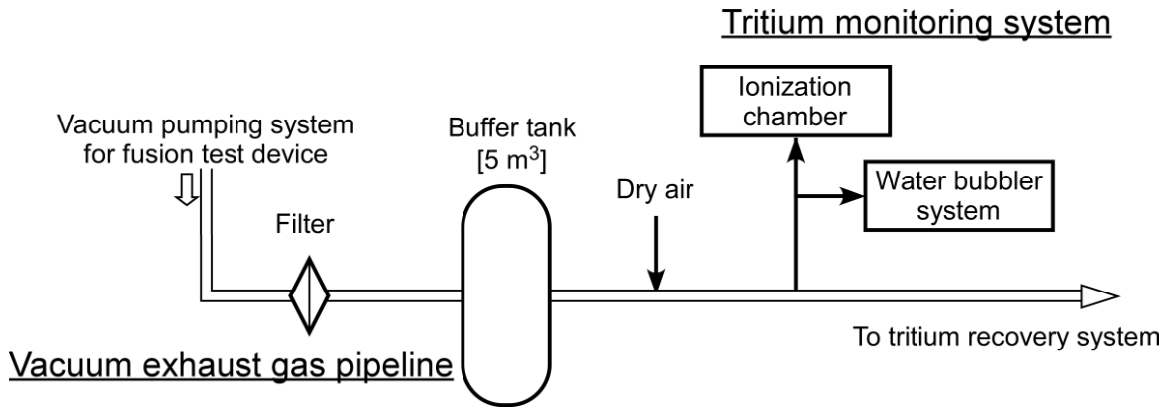
116 Ionization chamber for cross-checking the water bubbler system

117 To validate the tritium concentration measured by the proposed water bubbler system,  
118 an ionization chamber (Y221G0300, Ohkura Electric Co., Ltd.) was used. The volume of  
119 the aluminum ionization chamber was  $0.01 \text{ m}^3$  and the operation pressure was 0.098 MPa  
120 (G). The sampling gas flow rate was  $0.01 \text{ m}^3/\text{min}$ . The specification for the tritium  
121 detection limit is about  $7 \times 10^{-3} \text{ Bq/cm}^3$ . However, the practical background level measured  
122 in the exhaust gas was about  $4.4 \times 10^{-2} \text{ Bq/cm}^3$  because the background signal level was  
123 increased by interference from radionuclide such as radon gas.

#### 124 Tritium monitoring point

125 A schematic flow diagram of the tritium monitoring system is shown in Fig. 2. The  
126 water bubbler system and the ionization chamber were connected in parallel at the inlet of  
127 the exhaust detritiation system (EDS) [29]. The exhaust gas composition from the vacuum  
128 pumping system varied with the operation mode of the fusion test device. The exhaust gas  
129 from the fusion test device was purged with nitrogen gas at a flow rate of about  $4 \text{ Nm}^3/\text{h}$   
130 to prevent hydrogen explosions in the exhaust gas pipeline, and the exhaust gas did not  
131 contain oxygen. Thus, the tritium monitoring system was installed downstream of the dry  
132 air supply because oxygen gas must be added to oxidize the hydrogen and hydrocarbons.  
133 The addition of dry air contributed to the stable operation of the ionization chamber, which  
134 was designed for the tritium in air.

135 The water bubbler system began operating on Monday and accumulated samples for  
136 168 h. The ionization chamber operated continuously during the period in which the plasma  
137 experiments were performed. The tritium monitoring was performed during a deuterium  
138 plasma experiment, which produced a small amount of tritium.



139

140 **Fig. 2** Schematic diagram of the tritium monitoring point at the inlet of the EDS.

141 **Results and discussion**

142 **Catalytic oxidation under wet conditions**

143 When the sampling gas was passed through the water bubbler, water evaporated and  
 144 the sampling gas was humidified. Moisture degrades the oxidation performance of the  
 145 catalyst, and to prevent deterioration and the contamination of the tritiated water vapor, we  
 146 used a hydrophobic platinum catalyst, H1P, in the hydrogen oxidation reactor. The catalytic  
 147 oxidation performance using a small sample was evaluated under wet conditions by using  
 148 a fixed-bed catalyst flow reactor. The detailed specifications and the flow diagram of  
 149 apparatus have been described elsewhere [27]. The volume of the test sample and the  
 150 volume velocity were  $4.4 \text{ cm}^3$  and  $3.0 \text{ s}^{-1}$ , respectively. The humidity was set at dew points  
 151 of 278 and 283 K and at less than 253 K for dry conditions. The dew-point dependency of  
 152 the catalytic performance for 0.2% hydrogen is shown in Fig. 3. Here, the conversion rate  
 153 of oxidation,  $C$  (%), and the volume velocity,  $S_v$  ( $\text{s}^{-1}$ ), are defined by

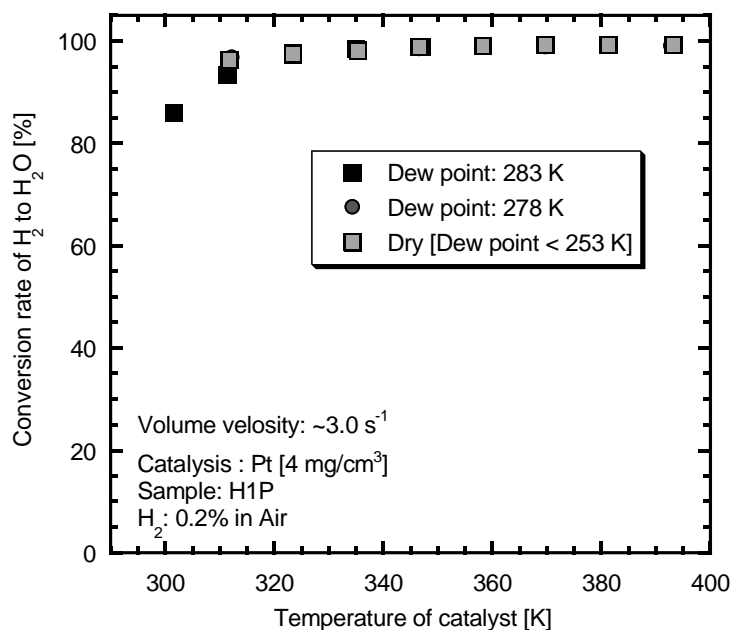
154 
$$C (\%) = \left(1 - \frac{C_{out}}{C_{in}}\right) \times 100, \quad (1)$$

155 
$$S_v (\text{s}^{-1}) = \frac{F}{V_{cat}}, \quad (2)$$

156 where  $C_{in}$  and  $C_{out}$  are the gas concentration at the inlet and the outlet of the catalyst bed,  
 157 respectively,  $V_{cat}$  is the catalyst volume, and  $F$  is the volume flow rate. The hydrogen



158 conversion rate was more than 99% at 353 K despite the wet conditions. Because the  
 159 volume velocity for the test operation was about two orders of magnitude larger than the  
 160 practical operation conditions, the hydrogen oxidation performance of H1P was sufficient  
 161 above 373 K. The dew point would be less than 283 K under practical conditions because  
 162 the water bubbler was installed in the incubator controlled at 275 K. Thus, the H1P catalyst  
 163 was suitable as the hydrogen oxidation catalyst in the water bubbler system at an operating  
 164 temperature of 373 K. On the other hand, it is known that the methane is not oxidized by a  
 165 platinum catalyst at the range of less than 450 K [27, 28]. Tritiated hydrocarbons would  
 166 not be converted to water vapor on the H1P at the range of less than 373K.



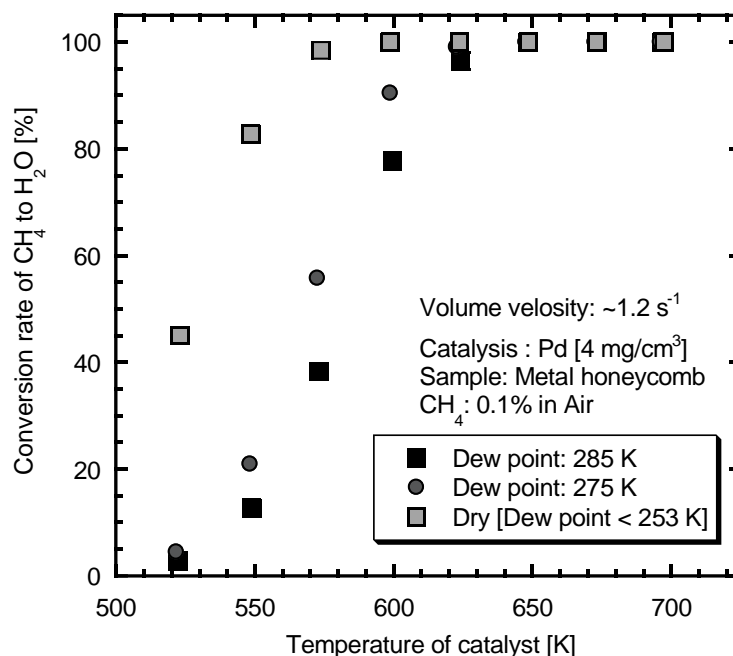
167

168 **Fig. 3** Effects of moisture on hydrogen gas oxidation reaction rate with H1P.

169

170 Figure 4 shows the performance of the metal honeycomb-supported palladium catalyst  
 171 for methane oxidation under wet conditions. In this oxidation performance test, the  
 172 methane was chosen as typical hydrocarbons, because the other hydrocarbons would be a  
 173 minority in the present plasma experimental condition [30]. The catalyst volume and the  
 174 volume velocity were 3.5 cm<sup>3</sup> and 1.2 s<sup>-1</sup>, respectively. The methane concentration was set  
 175 to 0.1%. The humidity was set at dew points of 275 and 285 K and at less than 253 K for  
 176 dry conditions. The methane oxidation performance was affected considerably by the water  
 177 vapor below 623 K. However, a methane conversion rate of 100% was achieved above 643

178 K. Because the volume velocity for the test operation was about one order of magnitude  
179 larger than the practical operation conditions, the metal honeycomb-supported palladium  
180 catalyst was used as the methane oxidation catalyst in the water bubbler system at an  
181 operating temperature of 673 K.



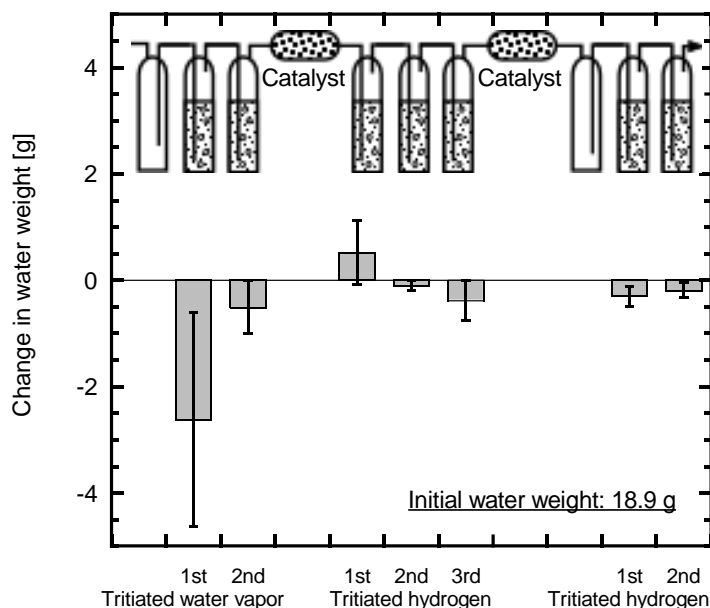
182

183 **Fig. 4** Effects of moisture in the process gas on methane gas oxidation reaction rate by  
184 the metal honeycomb-supported palladium catalyst.

### 185 Mass balance in the water bubbler system

186 Part of the water in the bubbler evaporates with the sampling gas. Thus, the sample  
187 water weight varies with sampling time. Figure 5 shows the variation of sample water  
188 weight in the bubbler after sampling. The average initial water weight in each bubbler was  
189 18.9 g. The sampling time was 1 week, but the flow rate varied in the range of 30 to 200  
190 cm<sup>3</sup>/min because tritium concentration changed by the operation of the LHD. The first  
191 bubbler water was evaporated by dry sample gas at a dew point below 243 K and the  
192 decrease in the water weight was  $2.6 \pm 2.0$  g. However, the decrease in the water weight in  
193 the second bubbler was only  $0.5 \pm 0.5$  g because the sampling gas after the first bubbler  
194 reached saturation at the incubator temperature. In contrast, the water weight in the first  
195 bubbler after the hydrogen oxidation reactor increased by  $0.5 \pm 0.5$  g. The hydrogen

196 concentration in the exhaust gas varied and increased in the range of 0.5% with the  
 197 operation conditions. Thus, the hydrogen gas in the sample gas was oxidized by the reactor,  
 198 and then the sample gas with saturated water vapor condensed in the bubbler at the  
 199 incubator temperature. After the third bubbler, the decreases in water weight in the bubblers  
 200 were less than 0.4 g.



201

**Fig. 5** Variation of bubbler water weight after sampling.

202

203

204 The tritium count rates in the water samples measured by a liquid scintillation counter  
 205 are summarized in Table 1. The collection efficiency, EFF, is defined as

206 
$$\text{EFF (\%)} = \left(1 - \frac{A_{nth}}{A_{1st}}\right) \times 100, \quad n: 2, 3 \quad (3)$$

207 where,  $A_{1st}$  and  $A_{nth}$  are the tritium count rate in the first and  $n$ th water bubbler, respectively.

208 When the sampling flow rate was 30 cm<sup>3</sup>/min, the average collection efficiency per  
 209 bubbler was 95.7% for tritiated water vapor and 96.9% for tritiated hydrocarbon. Although  
 210 more than 2.6 g of water in the first bubbler evaporated, the evaporated water vapor  
 211 containing tritium was collected in the second bubbler. In this case, tritium in the sample  
 212 gas was collected in the series of water bubblers with a collection efficiency of more than  
 213 99.9%. Thus, the effect of the tritium count rate in the downstream water sample was  
 214 several counts per minute according to Table 1. Since a part of tritium in the upstream

215 water bubbler transports and affects to the downstream water sampler, it is desirable that  
 216 the tritium count rate in the last water sample of each series of the bubbler is as low as  
 217 possible. When the sampling flow rate was 50 cm<sup>3</sup>/min, the average collection efficiency  
 218 via two bubblers was 99.7% for tritiated water vapor. The collection efficiency at a flow  
 219 rate of 50 cm<sup>3</sup>/min was similar to that at 30 cm<sup>3</sup>/min. However, when the sampling flow  
 220 rate was more than 100 cm<sup>3</sup>/min, the collection efficiencies via one and two bubblers were  
 221 90% and 98%, respectively. In these cases, the effect of the tritium count in the downstream  
 222 water sample would be more than several tens or hundreds of counts per minute. Thus, an  
 223 operation flow rate of less than 50 cm<sup>3</sup>/min was suitable for the tritium sampling using this  
 224 water bubbler volume. When the operation flow rate exceeds 50 cm<sup>3</sup>/min, the correction  
 225 for collection efficiency is applied based on the data in Table 1.

226 **Table 1** Tritium count rate in the water samples.

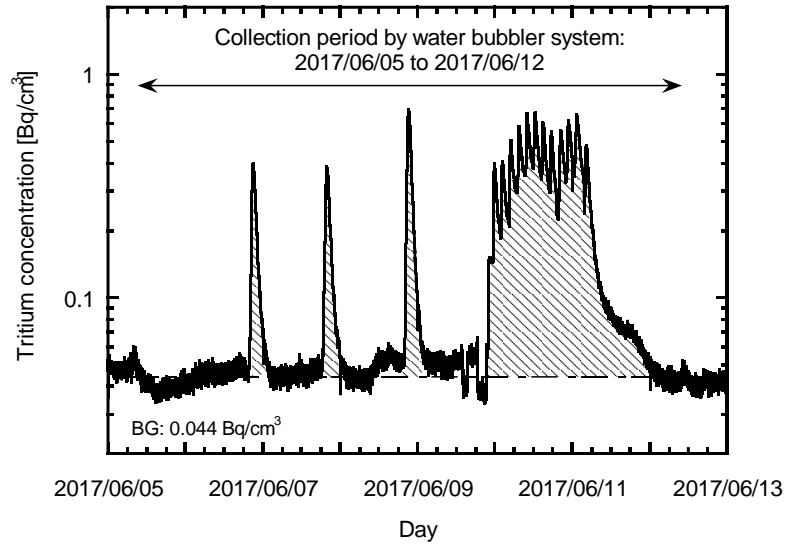
| Flow rate<br>[cm <sup>3</sup> /min] | Sampling<br>time [min] | Collection<br>sample gas<br>volume<br>[m <sup>3</sup> ] | Count rate by a liquid scintillation counter [cpm: counts per minute] |                       |                |  |                        |                |                |  |                       |                |  |
|-------------------------------------|------------------------|---|---|-----------------------|----------------|--|------------------------|----------------|----------------|--|-----------------------|----------------|--|
|                                     |                        |   | Background  | Tritiated water vapor |                |  | Tritiated hydrogen gas |                |                |  | Tritiated hydrocarbon |                |  |
|                                     |                        |   |   | 1st<br>bubbler        | 2nd<br>bubbler | Collection<br>efficiency<br>1st => 2nd | 1st<br>bubbler         | 2nd<br>bubbler | 3rd<br>bubbler | Collection<br>efficiency<br>1st => 3rd | 1st<br>bubbler        | 2nd<br>bubbler | Collection<br>efficiency<br>1st => 2nd |
| 30                                  | 9997                   | 0.299   | 2.9   | 368.7                 | 14.9           | 96.8%                                  | 155,679.0              | 3360.5         | 106.0          | 99.9%                                  | 5072.0                | 116.9          | 97.8%                                  |
| 30                                  | 10,160                 | 0.304   | 3.7   | 368.1                 | 25.0           | 94.5%                                  | 160,483.9              | 6241.8         | 211.6          | 99.9%                                  | 5409.4                | 207.8          | 96.4%                                  |
| 30                                  | 10,065                 | 0.301   | 3.5   | 299.9                 | 19.0           | 95.0%                                  | 170,202.6              | 4779.3         | 167.8          | 99.9%                                  | 7639.5                | 156.5          | 98.0%                                  |
| 30                                  | 10,065                 | 0.301   | 3.3   | 448.0                 | 20.6           | 96.2%                                  | 219,000.7              | 6961.9         | 176.0          | 99.9%                                  | 7237.8                | 264.4          | 96.5%                                  |
| 30                                  | 10,065                 | 0.301   | 3.4   | 460.9                 | 23.1           | 95.9%                                  | 122,994.5              | 5151.5         | 221.9          | 99.8%                                  | 4137.9                | 189.4          | 95.7%                                  |
|                                     |                        |   |   | Average               |                | 95.7%                                  |                        | Average        |                | 99.9%                                  |                       | Average        | 96.9%                                  |
| 50                                  | 10,136                 | 0.506   | 3.8   | 268.2                 | 11.6           | 97.1%                                  | 25,040.8               | 1108.2         | 42.5           | 99.9%                                  | 728.6                 | 67.4           | 91.9%                                  |
| 50                                  | 10,065                 | 0.503   | 3.4   | 163.3                 | 14.7           | 93.4%                                  | 30,026.6               | 817.0          | 73.9           | 99.8%                                  | 1510.8                | 28.5           | 98.4%                                  |
| 50                                  | 10,055                 | 0.502   | 3.1   | 519.2                 | 27.5           | 95.5%                                  | 62,436.4               | 2630.6         | 135.5          | 99.8%                                  | 1625.1                | 62.6           | 96.5%                                  |
| 50                                  | 9961                   | 0.498   | 5.6   | 520.0                 | 43.8           | 93.1%                                  | 47,807.0               | 2243.7         | 132.7          | 99.7%                                  | 839.8                 | 65.6           | 93.3%                                  |
| 50                                  | 10,062                 | 0.503   | 3.2   | 1266.9                | 56.2           | 96.0%                                  | 17,845.6               | 1585.3         | 111.9          | 99.4%                                  | 919.4                 | 77.3           | 92.5%                                  |
|                                     |                        |   |   | Average               |                | 95.0%                                  |                        | Average        |                | 99.7%                                  |                       | Average        | 94.5%                                  |
| 100                                 | 10,054                 | 1.01  | 3.2   | 163.4                 | 19.6           | 90.7%                                  | 44,149.8               | 4633.4         | 856.2          | 98.3%                                  | 1257.6                | 146.9          | 89.7%                                  |
| 200                                 | 10,064                 | 2.01  | 5.0   | 393.2                 | 46.4           | 90.4%                                  | 84,666.4               | 16633.7        | 4640.8         | 95.6%                                  | 3269.1                | 501.7          | 86.8%                                  |

227 Determination of tritium activity by the water bubbler system

228 To determine the tritium concentration measured by the water bubbler system, the  
229 average tritium concentration measured by the ionization chamber was compared with the  
230 tritium monitoring results from the water bubbler system. The typical variation of tritium  
231 concentration during plasma operation measured by the ionization chamber over 1 week is  
232 shown in Fig. 6. The background level of the ionization chamber was about 0.044 Bq/cm<sup>3</sup>.  
233 Several tritium concentration peaks were observed in the exhaust gas depending on the  
234 experimental operation. Thus, the average tritium concentration measured by the ionization  
235 chamber in a week,  $A_{ave}$ , was calculated by

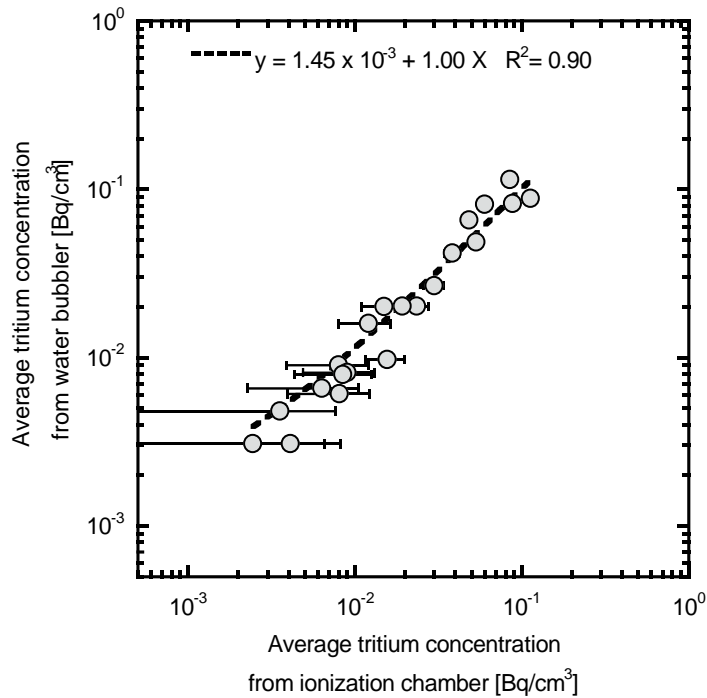
$$\begin{aligned} 236 \quad A_{ave} &= \frac{\sum_i \int_{t'}^{t''} F \times a_i(t) dt}{\int_0^{1week} F dt} \\ 237 \quad &= \frac{\sum_i A_i}{V}, \end{aligned} \quad (4)$$

238 where  $F$  is the constant process gas flow rate of 20 Nm<sup>3</sup>/h at the monitoring point in the  
239 EDS,  $a_i(t)$  is the tritium concentration of each peak signal subtracting the background signal,  
240  $i$  is the number of peaks,  $t'$  and  $t''$  are the times of the start and end of each tritium peak  
241 signal,  $A_i$  is the total amount of tritium in each peak, and  $V$  is the total process gas volume  
242 in the EDS in a week. The relationship between the average concentrations measured by  
243 the ionization chamber and water bubbler system is shown in Fig. 7. The average tritium  
244 concentration measured by the water bubbler system was the total tritium concentration of  
245 all chemical forms. The dashed line shows the linear regression curve. The gradient of the  
246 linear regression is almost unity and the coefficient of determination,  $R^2$ , is 0.90. The  
247 measurements suggest that there is a correlation between the ionization chamber and water  
248 bubbler system. Thus, the performance of the water bubbler system was verified and the  
249 tritium activity in the exhaust gas from LHD could be determined by the water bubbler  
250 system.



251  
252  
253  
254

**Fig. 6** Example of the variation of tritium concentration measured by the ionization chamber over 1 week at the EDS inlet. The hatched area shows the tritium signal with the background signal subtracted.



255

256  
257

**Fig. 7** Relationship between the average tritium concentrations measured by the ionization chamber and water bubbler system.

258 Observation of tritium chemical forms in the exhaust gas

259 Various hydrogen isotope compounds are produced in fusion test devices by plasma  
 260 surface interactions with materials, such as chemical erosion [31]. For example, various  
 261 deuterated hydrocarbons were formed by chemical erosion and observed in the exhaust gas  
 262 during deuterium plasma operation of the JET and JT-60U fusion test devices because the  
 263 plasma facing components are made of carbon [23, 32-34]. In this study, we observed the  
 264 exhaust gas from LHD, in which the first wall is stainless steel and the diverter tiles are  
 265 carbon. Tritiated hydrocarbons were detected by the proposed water bubbler system in the  
 266 exhaust gas during plasma experiments. The ratio of tritium chemical forms in the exhaust  
 267 gas from LHD is compared with data from JT-60U [21-24] in Table 2. The length of  
 268 collection was from several hours or 1 day in JT-60U to 1 week in LHD. The operation  
 269 modes were deuterium plasma experiments or glow discharge cleaning with hydrogen or  
 270 helium. In LHD and JT-60U, the main tritium chemical form was tritiated hydrogen gas  
 271 because the working gas was mainly hydrogen isotope gas, H<sub>2</sub> or D<sub>2</sub>. Tritiated water vapor  
 272 was less than 1% of the tritium in the exhaust gas. Tritiated hydrocarbons were several  
 273 percent of the tritium in the exhaust gas from both JT-60U and LHD. The ratio of tritiated  
 274 hydrocarbons from JT-60U was about twice that from LHD. The factors causing the  
 275 difference between JT-60U and LHD are the operating conditions, such as the number of  
 276 plasma shots and discharge duration; the first wall temperature; and the plasma parameters,  
 277 such as ion and electron temperature, and particle flux into the divertor tiles. Furthermore,  
 278 because the plasma-facing components in JT-60U are all carbon-based materials, the  
 279 tritiated hydrocarbons were formed more easily than in LHD. The proposed water bubbler  
 280 system for distinguishing chemical forms would be useful in understanding the tritium  
 281 behavior in a fusion test device.

282 **Table 2** Comparison of the ratio of tritium chemical forms in the exhaust gas from JT-  
 283 60U and LHD measured by water bubbler or silica gel traps

| Device | Operation mode       | Tritiated water vapor [%] | Tritiated hydrogen gas [%]                               | Tritiated hydrocarbons [%] | Collection period | Reference |
|--------|----------------------|---------------------------|--|----------------------------|-------------------|-----------|
| JT-60U | Glow discharge       | <1                        | >99<br>(Tritiated hydrogen gas + Tritiated hydrocarbons) |                            | Several hours     | Ref. 21   |
|        | Glow discharge, etc. | 0.45                      | >99.5  | -                          | 1 day             | Ref. 22   |
|        |                      | 0.39*                     | -  | -                          |                   | Ref. 23   |

|     |                             |             |            |             |                                   |            |
|-----|-----------------------------|-------------|------------|-------------|-----------------------------------|------------|
|     |                             | 0.35*       | -          | -           |                                   |            |
|     | Deuterium plasma experiment | 0.32*       | -          | -           |                                   |            |
|     |                             | 0.3         | 93.3       | 6.4         |                                   | Ref. 24    |
|     |                             | 0.2         | 95.5       | 4.3         |                                   |            |
| LHD | Deuterium plasma experiment | 0.21 ± 0.01 | 96.7 ± 0.3 | 3.06 ± 0.06 | 1 week (2017/06/05 to 2017/06/12) | This study |

284 \* These data were measured by a silica gel trap system

## 285 **Conclusions**

286 We commissioned a tritium monitoring system using the water bubbler method to  
 287 distinguish tritium chemical forms in the exhaust gas from a fusion test device. The  
 288 sampling performance of the proposed water bubbler system was evaluated under actual  
 289 operating conditions. The oxidation performance of the catalysts for distinguishing  
 290 chemical forms was adequate at the operating temperature. The tritium collection  
 291 efficiency in a series of two water bubbler columns was more than 99.9% at a flow rate of  
 292 30 cm<sup>3</sup>/min. Thus, the effect of tritium in the downstream bubbler could be ignored under  
 293 these operating conditions. When the operation flow rate was more than 50 cm<sup>3</sup>/min, the  
 294 correction for collection efficiency was necessary.

295 The tritium activity measurement using the water bubbler system was compared with  
 296 the ionization chamber. The correlation between the water bubbler system and ionization  
 297 chamber measurements was confirmed and the tritium activity in the exhaust gas from  
 298 LHD could be determined by the water bubbler system. Furthermore, the proposed water  
 299 bubbler system distinguished the tritium chemical forms. The tritium chemical forms in  
 300 the exhaust gas provide information about the tritium behavior in the fusion test device.

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304 **References**

- 305 1. Komori A, Yamada H, Imagawa S, Kaneko O, Kawahata K, Mutoh K, Ohyabu N,  
306 Takeiri Y, Ida K, Mito T, Nagayama Y, Sakakibara S, Sakamoto R, Shimozuma T,  
307 Watanabe KY, Motojima O, LHD Experiment Group (2010) Goal and achievements  
308 of Large Helical Device project. *Fusion Sci Technol* 58:1-11
- 309 2. Takeiri Y (2018) Prospect towards steady-state helical fusion reactor based on progress  
310 of LHD project entering the deuterium experiment phase, *IEEE Trans Plasma Sci*  
311 46:1141-1148
- 312 3. Osakabe M, Takeiri Y, Morisaki T, Motojima G, Ogawa K, Isobe M, Tanaka M,  
313 Murakami S, Shimizu A, Nagaoka K, Takahashi H, Nagasaki K, Takahashi H, Fujita  
314 T, Oya Y, Sakamoto M, Ueda Y, Akiyama T, Kasahara H, Sakakibara S, Sakamoto R,  
315 Tokitani M, Yamada H, Yokoyama M, Yoshimura Y and LHD Experiment Group  
316 (2017) Current status of Large Helical Device and its prospect for deuterium  
317 experiment. *Fusion Sci Technol* 72:199-210
- 318 4. IAEA (1991) SAFE HANDLING OF TRITIUM: Review of Data and Experience.  
319 IAEA TECHNICAL REPORTS SERIES No. 324, VIENNA
- 320 5. DOE HANDBOOK (2009) Tritium handling and safe storage. DOE-HDBK-1129-  
321 2008, U.S. Department of Energy, Washington, D.C. 20585
- 322 6. Osborne RV (1974) Sampling for tritiated water vapour. Proceedings of the Third  
323 International Congress of the International Radiation Protection Association,  
324 Washington D.C., 1973, CONF-730907-P2:1428-1433
- 325 7. Shank KE, Easterly CE (1976), Tritium instrumentation for a fusion reactor power  
326 plant. ORNL/TM-5344
- 327 8. Östlund HG, Mason AS (1974), Atmospheric HT and HTO, *Tellus* 26:91-102
- 328 9. Okai T, Takashima Y (1989), Analytical method for atmospheric tritium with a  
329 portable tritium sampling system, *J Radioanal Nucl Chem* 130:399-407
- 330 10. Otlet RL, Walker AJ, Caldwell-Nichols CJ (1992) Practical environmental, working  
331 area and stack discharge samplers, passive and dynamic, for measurement of tritium as  
332 HTO and HT, *Fusion Technol* 21:550-555

- 333 11. Wood MJ, McElroy RGC, Surette RA, Brown RM (1993), Tritium sampling and  
334 measurement, *Health Phys* 65(6):610-627
- 335 12. Munyon WJ, Reilly DW, Webb J (2008) AGHCF gaseous-effluent tritium sampling  
336 system: design considerations and performance testing results. *Health Phys* 94(1):75-  
337 84
- 338 13. SDEC France, MARC 7000 Tritium sampler. [http://radioprotection.sdec-](http://radioprotection.sdec-france.com/tritium-sampler-4-vials-marc-7000.html)  
339 [france.com/tritium-sampler-4-vials-marc-7000.html](http://radioprotection.sdec-france.com/tritium-sampler-4-vials-marc-7000.html). Accessed 2 Jul 2018
- 340 14. Overhoff Technology, TASC(Tritium in Air Sample Collector)-HTO-HT.  
341 <http://www.overhoff.com/uploads/Chapter5.pdf>. Accessed 2 Jul 2018
- 342 15. RCD Lockinge, Dynamic HTO/HT Discriminating Dry Bed Absorber Sampler.  
343 <http://rcd-lockinge.co.uk/equipment.htm>. Accessed 2 Jul 2018
- 344 16. Uda T, Sugiyama T, Tanaka M, Munakata K, Momoshima N (2006) Developments of  
345 gaseous water, hydrogen and methane sampling system for environmental tritium  
346 monitoring. *Fusion Eng Des* 81:1385–1390
- 347 17. Tanaka M, Kato H, Yamamoto Y, Iwata C (2017) Development of an active tritium  
348 sampler for discriminating chemical forms without the use of combustion gases in a  
349 fusion test facility. *Appl Radiat Isotop* 125:53-59
- 350 18. Duda JM, Goff PL, Leblois Y, Ponsard S (2018) Efficiencies of Tritium ( $^3\text{H}$ ) bubbling  
351 systems. *J Environ Radioact* 189:236-249.
- 352 19. Masaki K, Yagyū J, Arai T, Kaminaga A, Kodama K, Miya N, Ando T, Hiratsuka H,  
353 Saidoh M (2002) Wall conditioning and experience of the carbon-based first wall in  
354 JT-60U, *Fusion Sci Technol* 42:386-395
- 355 20. Isobe K, Nakamura H, Kaminaga A, Higashijima S, Nishi M, Konishi S, Nishikawa M,  
356 Tanabe T (2005) Tritium release behavior from JT-60U vacuum vessel during air  
357 exposure phase and wall conditioning phase. *Fusion Sci Technol* 48:302-305
- 358 21. Nakamura H, Higashijima S, Isobe K, Kaminaga A, Horikawa T, Kubo H, Miya N,  
359 Nishi M, Konishi S, Tanabe T (2004) Application of glow discharges for tritium  
360 removal from JT-60U vacuum vessel. *Fusion Eng Des* 70:163–173
- 361 22. Miya N, Nemoto M, Toyoshima N (1994) Tritium release from JT-60U vacuum vessel  
362 following high-power heated deuterium operations. *Fusion Technol* 26:507-511

- 363 23. Kaminaga A, Horikawa T, Nakamura H, Isobe K, Higashijima S, Arai T, Miya N, Nishi  
364 M, Tanabe T (2003) Analysis of exhausted gas in JT-60 deuterium operation. Proc. of  
365 20th IEEE/NPSS Symposium on Fusion Engineering, San Diego, CA, Oct. 14–17,  
366 2003
- 367 24. Kaminaga A, Nakamura H, Isobe K, Arai T (2005) Exhaust gas monitoring device of  
368 nuclear fusion experimental device. Proc. of Tech2005, Osaka, Japan, Mar. 3-4, 2005,  
369 83 [in Japanese].
- 370 25. ICRP (1990) Recommendations of the International Commission on Radiological  
371 Protection. ICRP Publication 60. Ann. ICRP 21(1-3)
- 372 26. Iwai Y, Sato K, Taniuchi J, Noguchi H, Kubo H, Harada N, Oshima Y, Yamashita T  
373 (2011) Room-temperature reactor packed with hydrophobic catalysts for the oxidation  
374 of hydrogen isotopes released in a nuclear facility. J Nucl Sci Technol 48(8):1184-1192
- 375 27. Uda T, Tanaka M, Munakata K (2008) Characteristics of honeycomb catalysts for  
376 oxidation of tritiated hydrogen and methane gases. Fusion Eng Des 83:1715–1720
- 377 28. Tanaka M, Uda T, Shinozaki Y, Munakata K (2009) Hydrogen and methane oxidation  
378 performances of hybrid honeycomb catalyst for a tritium removal system. Fusion Eng  
379 Des 84: 1818–1822
- 380 29. Tanaka M, Suzuki N, Kato H, Kondo T, Yokosawa M, Kawamata T, Ikeda M, Meguro  
381 T, Tanaka T, Sono K (2018) Design and commissioning of the exhaust detritiation  
382 system for the Large Helical Device. Fusion Eng Des 127:275–283
- 383 30. Isobe K, Nakamura H, Kaminaga A, Tsuzuki K, Higashijima S, Nishi M, Kobayashi  
384 Y, Konishi S (2006) Characterization of JT-60U exhaust gas during experimental  
385 operation. Fusion Eng Des 81:827-832
- 386 31. Roth J (2005) In: Clark EH, and Reiter DH (eds) Nuclear Fusion Research;  
387 Understanding Plasma-Surface Interactions. Springer, Berlin, Heidelberg
- 388 32. Grünhagen S, Brennan PD, Knipe S, Stagg R, Yorkshades J, JET-EFDA Contributors  
389 (2011) Analysis of hydrocarbons of the JET divertor cryogenic pump at the end of the  
390 carbon wall campaign using a micro gas chromatograph. Fusion Sci Technol  
391 60(3):931-936
- 392 33. Grünhagen Romanelli S, Brezinsek S, Butler B, Coad JP, Drenik A, Giroud C,  
393 Jachmich S, Keenan T, Kruezi U, Mozetic M, Oberkofler M, Parracho A, Romanelli

- 394 M, Smith R, Yorkshades J, JET-EFDA Contributors (2014) Gas analyses of the first  
395 complete JET cryopump regeneration with ITER-like wall. Phys Scr T159:014068  
396 34. Nakano T, Kubo H, Higashijima S, Asakura N, Takenaga H, Sugie T, Itami K (2002)  
397 Measurement of the chemical sputtering yields of CH<sub>4</sub>/CD<sub>4</sub> and C<sub>2</sub>H<sub>x</sub>/C<sub>2</sub>D<sub>x</sub> at the  
398 carbon divertor plates of JT-60U. Nucl Fusion 42:689–696