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

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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**

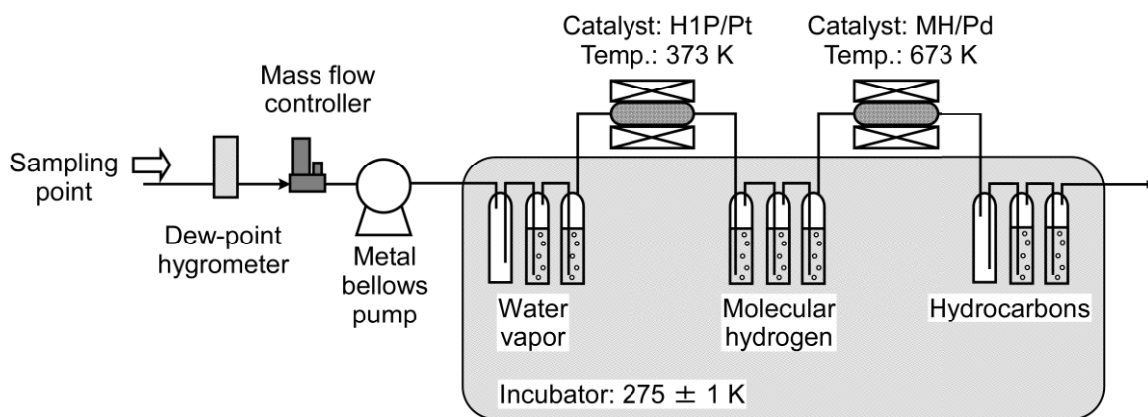
75 A flow diagram of the proposed water bubbler system is shown in Fig. 1. The water
76 bubbler system consisted of a dew-point hygrometer (Easidew Transmitter, Michell
77 Instruments Ltd.), a mass flow controller (8500MC, Kofloc), a metal bellows pump (MB-
78 21, IBS Inc.), a series of water bubbler columns (080100-02, volume: 30 cm³, 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: $\geq 18.2 \text{ M}\Omega\cdot\text{cm}$, TOC: $\leq 5 \text{ ppb}$). 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 ± 1
85 K. The gas sampling was performed at a flow rate of less than 200 cm³/min for 1 week.
86 The total amount of air sampled was less than 2 m³.

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/m³, packed weight:
92 290 g, outside diameter [O.D.]: $\phi 40 \text{ mm}$, 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/cm³, O.D.: ϕ 26 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 cm³) from each of the bubblers was mixed
103 with liquid scintillator (10 cm³, Ultima-Gold LLT, Perkin Elmer Co., Ltd.) in 20 cm³
104 polyethylene vials. The background sample was prepared using deionized water in a 20
105 cm³ 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/cm³. Although the detection limit of the gaseous tritium
111 concentration depended on the sampling volume, it was less than 10⁻⁶ Bq/cm³.

112



113

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

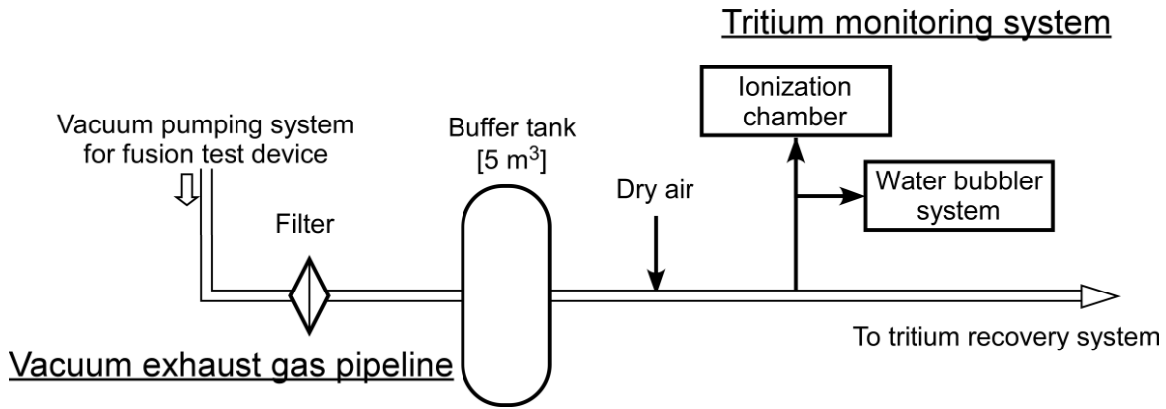
116 Ionization chamber for cross-checking the water bubbler system

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 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**

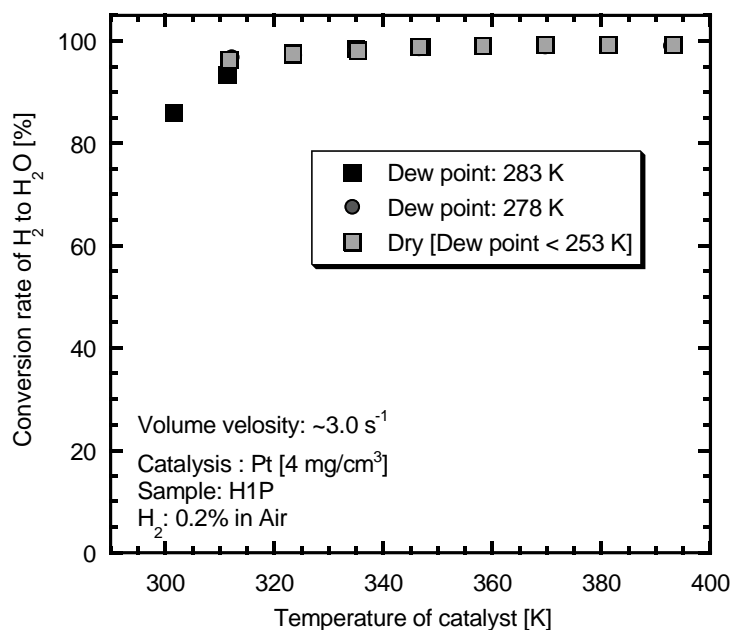
143 When the sampling gas was passed through the water bubbler, water evaporated and
 144 the sampling gas was humidified. Moisture degrades the oxidation performance of the
 145 catalyst, and to prevent deterioration and the contamination of the tritiated water vapor, we
 146 used a hydrophobic platinum 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 cm^3 and 3.0 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 (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.



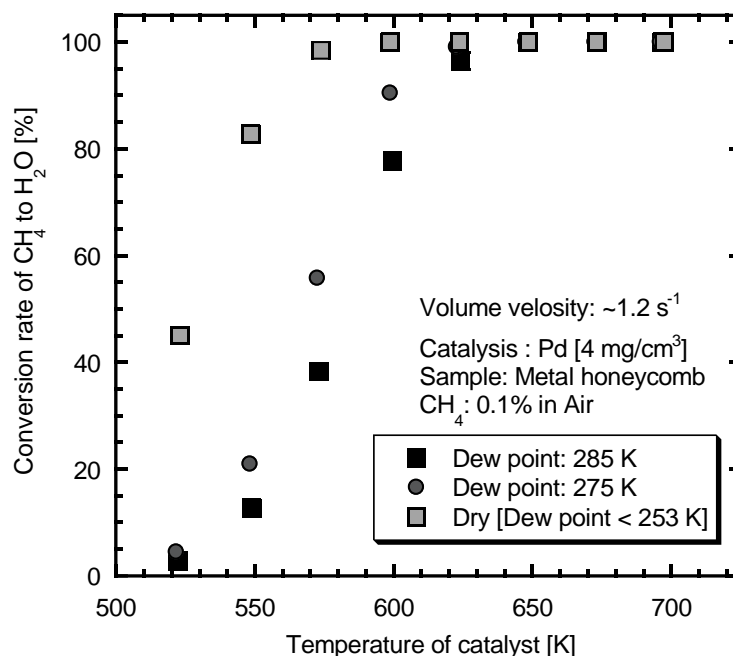
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168 **Fig. 3** Effects of moisture on hydrogen gas oxidation reaction rate with H1P.

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 cm³ and 1.2 s⁻¹, 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.



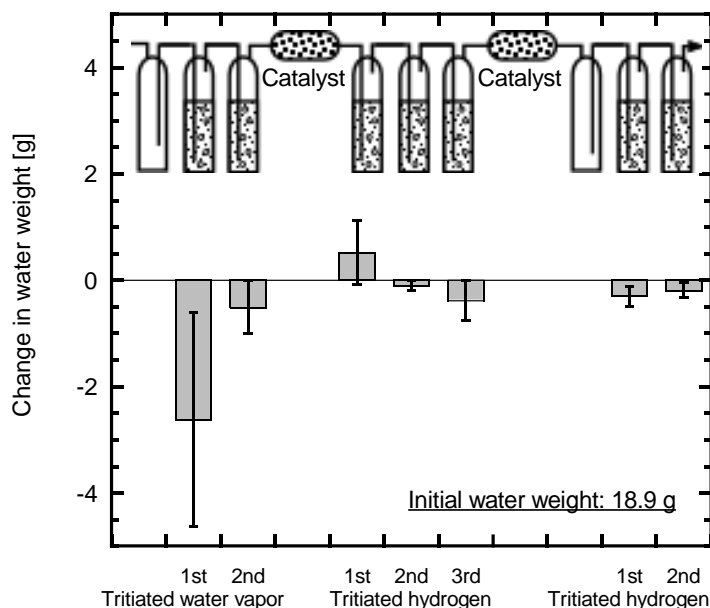
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183 **Fig. 4** Effects of moisture in the process gas on methane gas oxidation reaction rate by
184 the metal honeycomb-supported palladium catalyst.

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 cm³/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 ± 2.0 g. However, the decrease in the water weight in
193 the second bubbler was only 0.5 ± 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 ± 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

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

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 cm³/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 cm³/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 cm³/min was similar to that at 30 cm³/min. However, when the sampling flow
 220 rate was more than 100 cm³/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 cm³/min was suitable for the tritium sampling using this
 224 water bubbler volume. When the operation flow rate exceeds 50 cm³/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.

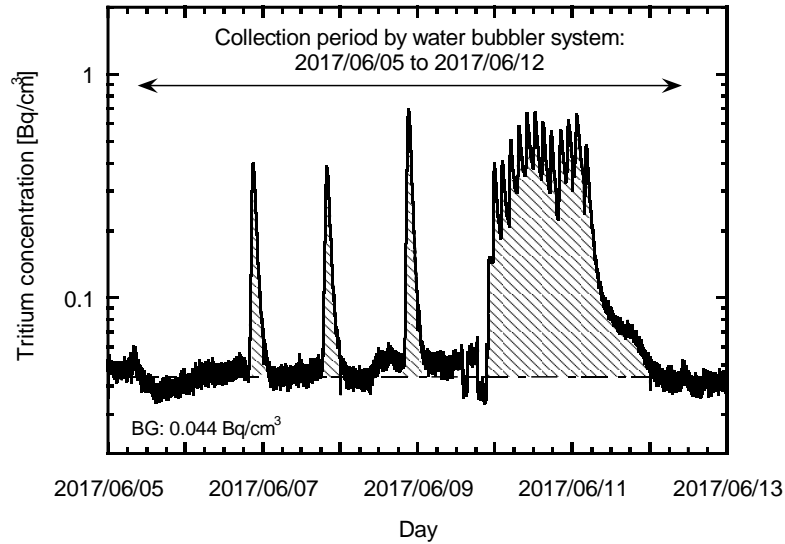
Flow rate [cm ³ /min]	Sampling time [min]	Collection sample gas volume [m ³]	Count rate by a liquid scintillation counter [cpm: counts per minute]										
			Background	Tritiated water vapor			Tritiated hydrogen gas				Tritiated hydrocarbon		
				1st bubbler	2nd bubbler	Collection efficiency 1st => 2nd	1st bubbler	2nd bubbler	3rd bubbler	Collection efficiency 1st => 3rd	1st bubbler	2nd bubbler	Collection efficiency 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/cm³.
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

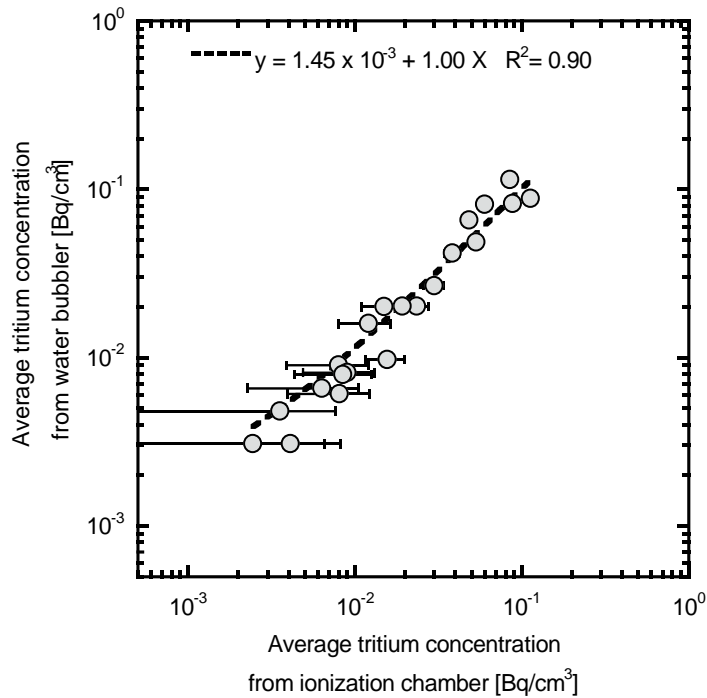
$$236 \quad A_{ave} = \frac{\sum_i \int_{t'}^{t''} F \times a_i(t) dt}{\int_0^{1week} F dt}$$
$$237 \quad = \frac{\sum_i A_i}{V}, \quad (4)$$

238 where F is the constant process gas flow rate of 20 Nm³/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.



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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, H₂ or D₂. 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 (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 cm³/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 cm³/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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