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

1   **Title page**

2   Names of the authors: Masahiro Tanaka[1, 2], Hiromi Kato[1], Naoyuki Suzuki[1], Chie Iwata[1],

3   Naofumi Akata[1, 2]

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[1, 2], Hiromi Kato[1], Naoyuki Suzuki[1], Chie Iwata[1], Naofumi Akata[1, 2]

16   *[1]National Institute for Fusion Science, National Institutes of Natural Sciences, Oroshi-*
17   *cho, Toki, Gifu 509-5292, Japan*

18   *[2]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^3, 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 MΩ·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^3/min for 1 week.

86   The total amount of air sampled was less than 2 m^3.

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^3, packed weight:

92   290 g, outside diameter [O.D.]: ϕ40 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^3, 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^3) from each of the bubblers was mixed

103   with liquid scintillator (10 cm^3, Ultima-Gold LLT, Perkin Elmer Co., Ltd.) in 20 cm^3

104   polyethylene vials. The background sample was prepared using deionized water in a 20

105   cm^3 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^3. Although the detection limit of the gaseous tritium

111   concentration depended on the sampling volume, it was less than 10^{-6} Bq/cm^3.

112

113
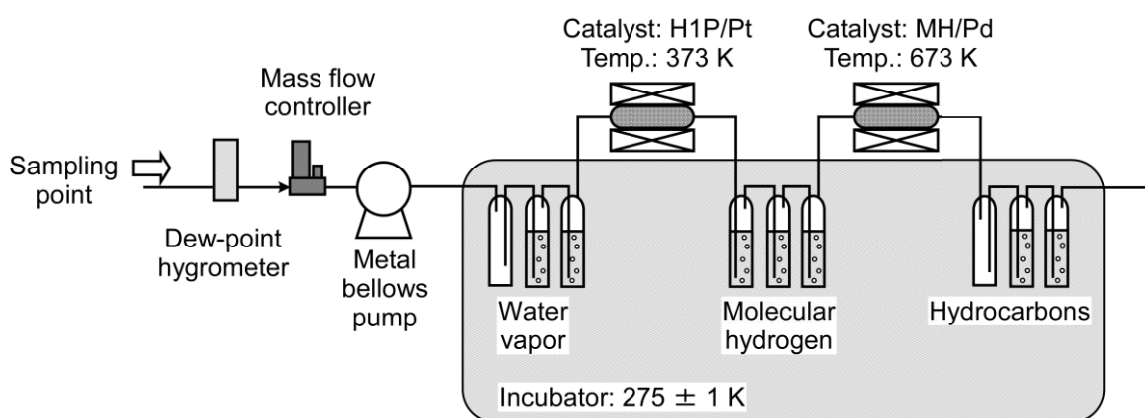
114   **Fig. 1** Flow diagram of the water bubbler system for distinguishing chemical forms of

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 m^3/min. The specification for the tritium

121   detection limit is about 7×10^{-3} Bq/cm^3. However, the practical background level measured

122   in the exhaust gas was about 4.4×10^{-2} 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 Nm^3/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
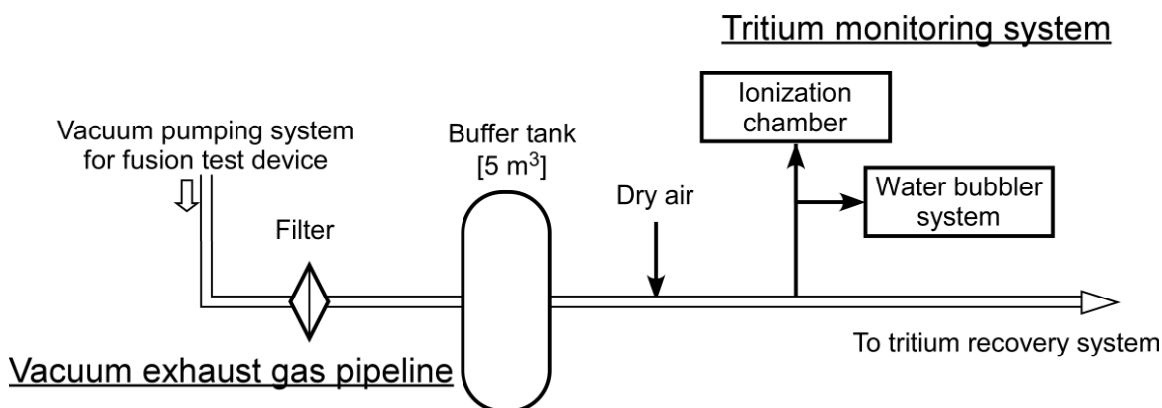
140 **Fig. 2** Schematic diagram of the tritium monitoring point at the inlet of the EDS.

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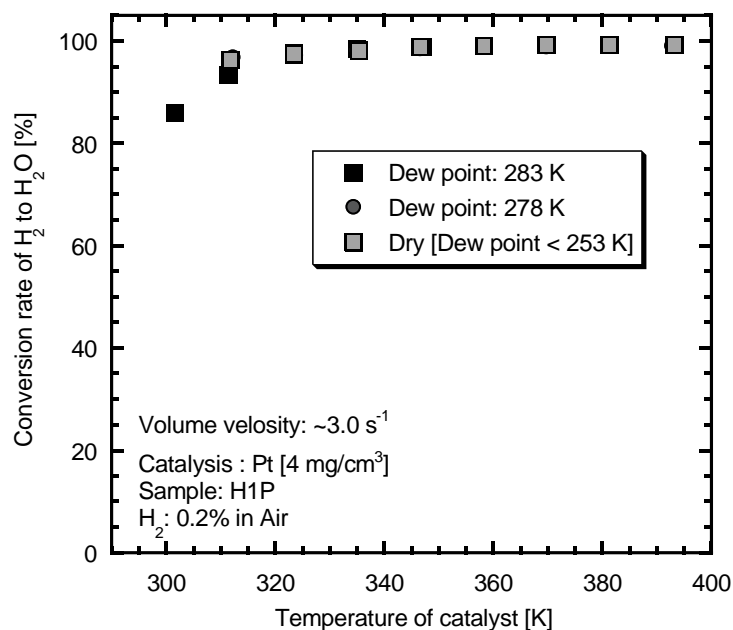
154 $$C\ (\%) = \left(1 - \frac{C_{out}}{C_{in}}\right) \times 100, \qquad\qquad (1)$$

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

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.

Legend inside figure:
- Dew point: 283 K
- Dew point: 278 K
- Dry [Dew point < 253 K]

Volume velosity: ~3.0 s^{-1}
Catalysis : Pt [4 mg/cm^3]
Sample: H1P
H$_2$: 0.2% in Air

Y-axis: Conversion rate of H$_2$ to H$_2$O [%]
X-axis: Temperature of catalyst [K]

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^3 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.

182
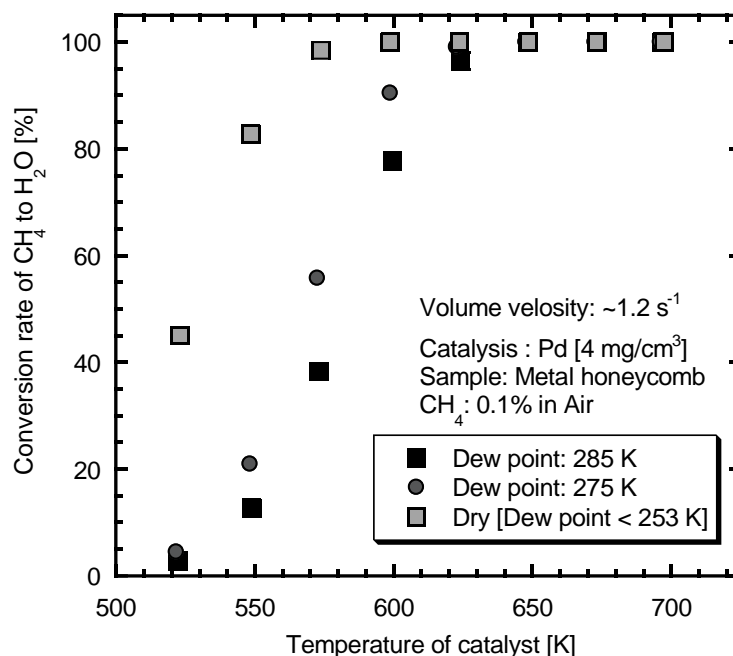
183   **Fig. 4** Effects of moisture in the process gas on methane gas oxidation reaction rate by
184   the metal honeycomb-supported palladium catalyst.

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^3/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
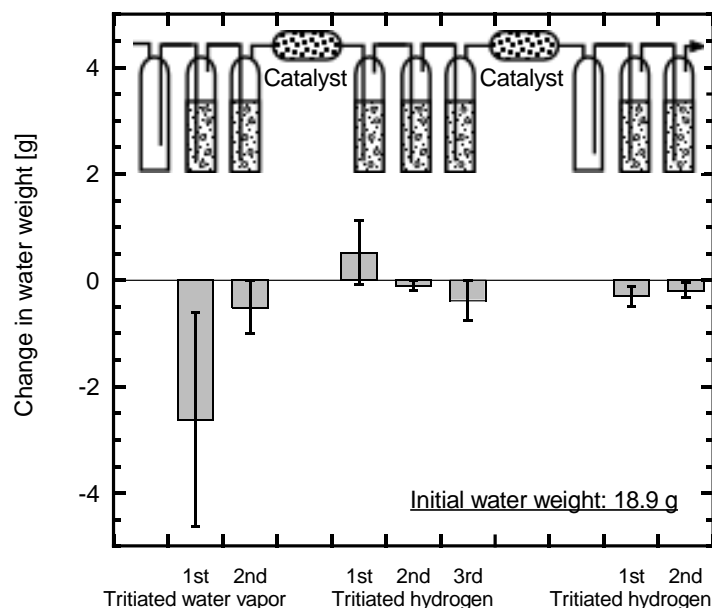
202   **Fig. 5** Variation of bubbler water weight after sampling.

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

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

208   When the sampling flow rate was 30 cm^3/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^3/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^3/min was similar to that at 30 cm^3/min. However, when the sampling flow

220   rate was more than 100 cm^3/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^3/min was suitable for the tritium sampling using this

224   water bubbler volume. When the operation flow rate exceeds 50 cm^3/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]	Background	Count rate by a liquid scintillation counter [cpm: counts per minute]										
				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%

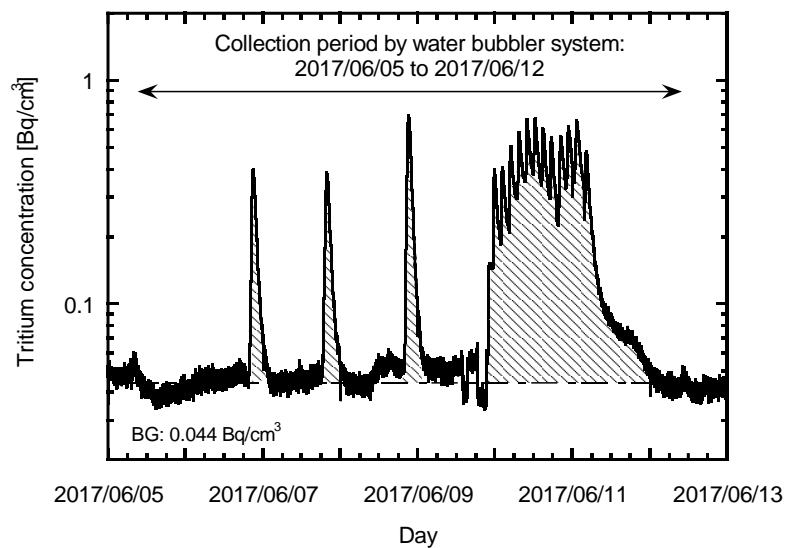227   Determination of tritium activity by the water bubbler system

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/cm^3.
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
$$A_{ave} = \frac{\sum_i \int_{t'}^{t''} F \times a_i(t) dt}{\int_0^{1week} F dt}$$

237
$$= \frac{\sum_i A_i}{V}, \qquad (4)$$

238   where F is the constant process gas flow rate of 20 Nm^3/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.

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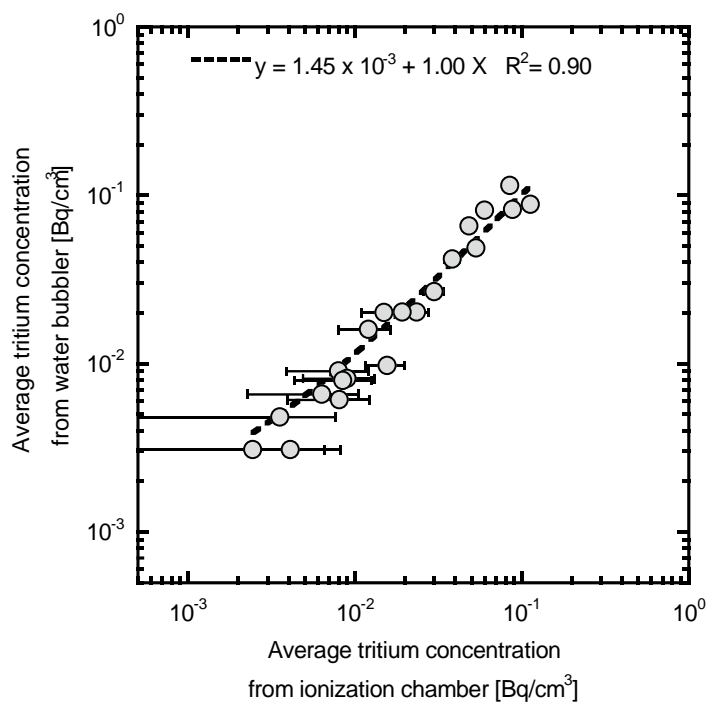
Fig. 7 Relationship between the average tritium concentrations measured by the ionization chamber and water bubbler system.

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_2 or D_2. 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

14

		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

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^3/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^3/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.

Acknowledgements

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-
339   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 (3H) bubbling
351   systems. J Environ Radioact 189:236-249.

352   19. Masaki K, Yagyu 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, Sonoi 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_4/CD_4 and C_2H_x/C_2D_x at the

398   carbon divertor plates of JT-60U. Nucl Fusion 42:689–696