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

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Abstract

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

Keywords

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

Introduction

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

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

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

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

Tritium monitoring apparatus and analytical methods

Water bubbler system

A flow diagram of the proposed water bubbler system is shown in Fig. 1. The water bubbler system consisted of a dew-point hygrometer (Easidew Transmitter, Michell Instruments Ltd.), a mass flow controller (8500MC, Kofloc), a metal bellows pump (MB-21, IBS Inc.), a series of water bubbler columns (080100-02, volume: 30 cm³, glass filter: P160, SIBATA SCIENTIFIC TECHNOLOGY LTD.), a two-stage oxidation reactor, and a low-temperature incubator (MIR-153, SANYO). Each bubbler contained about 19 g of deionized water produced by an ultrapure water system (Direct-Q UV, Merck Millipore, Resistivity at 298 K: ≥ 18.2 M Ω ·cm, TOC: ≤ 5 ppb). The water bubbler column was installed in the incubator to decrease water evaporation in the bubbler for long sampling times of more than 1 day. The operating temperature in the incubator was kept at 275 ± 1 K. The gas sampling was performed at a flow rate of less than 200 cm³/min for 1 week. The total amount of air sampled was less than 2 m³.

Tritium in the chemical form of water vapor was collected in the first bubbler series when sample gas containing tritium was passed through water. Then, the sample gas was passed through the low-temperature oxidation reactor to convert the tritiated hydrogen gas into tritiated water vapor. The low-temperature oxidation reactor was packed with a H1P hydrophobic platinum catalyst (Tanaka Kikinzoku Industry, Pt: 4 mg/m³, packed weight: 290 g, outside diameter [O.D.]: $\phi 40$ mm, and length: 300 mm) [26]. The hydrophobic catalyst was chosen to prevent the memory effect and the degradation of the hydrogen

oxidation performance under wet conditions at low temperature. The operation temperature of the oxidation reactor was kept around 373 K. The tritiated water vapor converted from tritiated hydrogen gas was collected in a second series of water bubbler columns. The residual tritium in the chemical form of tritiated hydrocarbons was then converted into tritiated water vapor by a palladium catalyst supported on a metal honeycomb (Tanaka Kikinzoku Industry, Pd: 4 mg/cm³, O.D.: ϕ 26 mm, length: 50 mm, cell density: 300 CPSI) [27]. The catalyst was heated to about 673 K. Finally, the tritiated water vapor converted from tritiated hydrocarbons was collected in the third series of water bubbler columns.

To determine the tritium activity, water (10 cm³) from each of the bubblers was mixed with liquid scintillator (10 cm³, Ultima-Gold LLT, Perkin Elmer Co., Ltd.) in 20 cm³ polyethylene vials. The background sample was prepared using deionized water in a 20 cm³ vial. The deionized water for the background sample was same as ultrapure water used in the bubbler column. After leaving these samples for several hours in a liquid scintillation counter (Tri-Carb 4910TR, Perkin Elmer Co. Ltd.), the tritium activity was determined for a total counting time of 50 min per sample. Counting efficiencies were determined by use of tritium standard solution (SRM 4361C, NIST, USA). The detection limit of the sample water was approximately 0.01 Bq/cm³. Although the detection limit of the gaseous tritium concentration depended on the sampling volume, it was less than 10⁻⁶ Bq/cm³.

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

Ionization chamber for cross-checking the water bubbler system

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

Tritium monitoring point

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

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

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

Results and discussion

Catalytic oxidation under wet conditions

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

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

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

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

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

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

Figure 4 shows the performance of the metal honeycomb-supported palladium catalyst for methane oxidation under wet conditions. In this oxidation performance test, the methane was chosen as typical hydrocarbons, because the other hydrocarbons would be a minority in the present plasma experimental condition [30]. The catalyst volume and the volume velocity were 3.5 cm³ and 1.2 s⁻¹, respectively. The methane concentration was set to 0.1%. The humidity was set at dew points of 275 and 285 K and at less than 253 K for dry conditions. The methane oxidation performance was affected considerably by the water vapor below 623 K. However, a methane conversion rate of 100% was achieved above 643

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

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

Mass balance in the water bubbler system

Part of the water in the bubbler evaporates with the sampling gas. Thus, the sample water weight varies with sampling time. Figure 5 shows the variation of sample water weight in the bubbler after sampling. The average initial water weight in each bubbler was 18.9 g. The sampling time was 1 week, but the flow rate varied in the range of 30 to 200 cm³/min because tritium concentration changed by the operation of the LHD. The first bubbler water was evaporated by dry sample gas at a dew point below 243 K and the decrease in the water weight was 2.6 ± 2.0 g. However, the decrease in the water weight in the second bubbler was only 0.5 ± 0.5 g because the sampling gas after the first bubbler reached saturation at the incubator temperature. In contrast, the water weight in the first bubbler after the hydrogen oxidation reactor increased by 0.5 ± 0.5 g. The hydrogen

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

Fig. 5 Variation of bubbler water weight after sampling.

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

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

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

When the sampling flow rate was 30 cm³/min, the average collection efficiency per bubbler was 95.7% for tritiated water vapor and 96.9% for tritiated hydrocarbon. Although more than 2.6 g of water in the first bubbler evaporated, the evaporated water vapor containing tritium was collected in the second bubbler. In this case, tritium in the sample gas was collected in the series of water bubblers with a collection efficiency of more than 99.9%. Thus, the effect of the tritium count rate in the downstream water sample was several counts per minute according to Table 1. Since a part of tritium in the upstream

water bubbler transports and affects to the downstream water sampler, it is desirable that the tritium count rate in the last water sample of each series of the bubbler is as low as possible. When the sampling flow rate was 50 cm³/min, the average collection efficiency via two bubblers was 99.7% for tritiated water vapor. The collection efficiency at a flow rate of 50 cm³/min was similar to that at 30 cm³/min. However, when the sampling flow rate was more than 100 cm³/min, the collection efficiencies via one and two bubblers were 90% and 98%, respectively. In these cases, the effect of the tritium count in the downstream water sample would be more than several tens or hundreds of counts per minute. Thus, an operation flow rate of less than 50 cm³/min was suitable for the tritium sampling using this water bubbler volume. When the operation flow rate exceeds 50 cm³/min, the correction for collection efficiency is applied based on the data in Table 1.

Table 1 Tritium count rate in the water samples.

Flow rate [cm³/min]	Sampling time [min]	Collection sample gas volume [m³]	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%	

Determination of tritium activity by the water bubbler system

To determine the tritium concentration measured by the water bubbler system, the average tritium concentration measured by the ionization chamber was compared with the tritium monitoring results from the water bubbler system. The typical variation of tritium concentration during plasma operation measured by the ionization chamber over 1 week is shown in Fig. 6. The background level of the ionization chamber was about 0.044 Bq/cm³. Several tritium concentration peaks were observed in the exhaust gas depending on the experimental operation. Thus, the average tritium concentration measured by the ionization chamber in a week, A_{ave} , was calculated by

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

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

where F is the constant process gas flow rate of 20 Nm³/h at the monitoring point in the EDS, $a_i(t)$ is the tritium concentration of each peak signal subtracting the background signal, i is the number of peaks, t' and t'' are the times of the start and end of each tritium peak signal, A_i is the total amount of tritium in each peak, and V is the total process gas volume in the EDS in a week. The relationship between the average concentrations measured by the ionization chamber and water bubbler system is shown in Fig. 7. The average tritium concentration measured by the water bubbler system was the total tritium concentration of all chemical forms. The dashed line shows the linear regression curve. The gradient of the linear regression is almost unity and the coefficient of determination, R^2 , is 0.90. The measurements suggest that there is a correlation between the ionization chamber and water bubbler system. Thus, the performance of the water bubbler system was verified and the tritium activity in the exhaust gas from LHD could be determined by the water bubbler 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

Various hydrogen isotope compounds are produced in fusion test devices by plasma surface interactions with materials, such as chemical erosion [31]. For example, various deuterated hydrocarbons were formed by chemical erosion and observed in the exhaust gas during deuterium plasma operation of the JET and JT-60U fusion test devices because the plasma facing components are made of carbon [23, 32-34]. In this study, we observed the exhaust gas from LHD, in which the first wall is stainless steel and the diverter tiles are carbon. Tritiated hydrocarbons were detected by the proposed water bubbler system in the exhaust gas during plasma experiments. The ratio of tritium chemical forms in the exhaust gas from LHD is compared with data from JT-60U [21-24] in Table 2. The length of collection was from several hours or 1 day in JT-60U to 1 week in LHD. The operation modes were deuterium plasma experiments or glow discharge cleaning with hydrogen or helium. In LHD and JT-60U, the main tritium chemical form was tritiated hydrogen gas because the working gas was mainly hydrogen isotope gas, H_2 or D_2 . Tritiated water vapor was less than 1% of the tritium in the exhaust gas. Tritiated hydrocarbons were several percent of the tritium in the exhaust gas from both JT-60U and LHD. The ratio of tritiated hydrocarbons from JT-60U was about twice that from LHD. The factors causing the difference between JT-60U and LHD are the operating conditions, such as the number of plasma shots and discharge duration; the first wall temperature; and the plasma parameters, such as ion and electron temperature, and particle flux into the divertor tiles. Furthermore, because the plasma-facing components in JT-60U are all carbon-based materials, the tritiated hydrocarbons were formed more easily than in LHD. The proposed water bubbler system for distinguishing chemical forms would be useful in understanding the tritium behavior in a fusion test device.

Table 2 Comparison of the ratio of tritium chemical forms in the exhaust gas from JT-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

	Deuterium plasma experiment	0.35*	-	-	Ref. 24	
		0.32*	-	-		
		0.3	93.3	6.4		
		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

* These data were measured by a silica gel trap system

Conclusions

We commissioned a tritium monitoring system using the water bubbler method to distinguish tritium chemical forms in the exhaust gas from a fusion test device. The sampling performance of the proposed water bubbler system was evaluated under actual operating conditions. The oxidation performance of the catalysts for distinguishing chemical forms was adequate at the operating temperature. The tritium collection efficiency in a series of two water bubbler columns was more than 99.9% at a flow rate of 30 cm³/min. Thus, the effect of tritium in the downstream bubbler could be ignored under these operating conditions. When the operation flow rate was more than 50 cm³/min, the correction for collection efficiency was necessary.

The tritium activity measurement using the water bubbler system was compared with the ionization chamber. The correlation between the water bubbler system and ionization chamber measurements was confirmed and the tritium activity in the exhaust gas from LHD could be determined by the water bubbler system. Furthermore, the proposed water bubbler system distinguished the tritium chemical forms. The tritium chemical forms in 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, Shimoizuma 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 (^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

23. Kaminaga A, Horikawa T, Nakamura H, Isobe K, Higashijima S, Arai T, Miya N, Nishi M, Tanabe T (2003) Analysis of exhausted gas in JT-60 deuterium operation. Proc. of 20th IEEE/NPSS Symposium on Fusion Engineering, San Diego, CA, Oct. 14–17, 2003
24. Kaminaga A, Nakamura H, Isobe K, Arai T (2005) Exhaust gas monitoring device of nuclear fusion experimental device. Proc. of Tech2005, Osaka, Japan, Mar. 3-4, 2005, 83 [in Japanese].
25. ICRP (1990) Recommendations of the International Commission on Radiological Protection. ICRP Publication 60. Ann. ICRP 21(1-3)
26. Iwai Y, Sato K, Taniuchi J, Noguchi H, Kubo H, Harada N, Oshima Y, Yamashita T (2011) Room-temperature reactor packed with hydrophobic catalysts for the oxidation of hydrogen isotopes released in a nuclear facility. J Nucl Sci Technol 48(8):1184-1192
27. Uda T, Tanaka M, Munakata K (2008) Characteristics of honeycomb catalysts for oxidation of tritiated hydrogen and methane gases. Fusion Eng Des 83:1715–1720
28. Tanaka M, Uda T, Shinozaki Y, Munakata K (2009) Hydrogen and methane oxidation performances of hybrid honeycomb catalyst for a tritium removal system. Fusion Eng Des 84: 1818–1822
29. Tanaka M, Suzuki N, Kato H, Kondo T, Yokosawa M, Kawamata T, Ikeda M, Meguro T, Tanaka T, Sono K (2018) Design and commissioning of the exhaust detritiation system for the Large Helical Device. Fusion Eng Des 127:275–283
30. Isobe K, Nakamura H, Kaminaga A, Tsuzuki K, Higashijima S, Nishi M, Kobayashi Y, Konishi S (2006) Characterization of JT-60U exhaust gas during experimental operation. Fusion Eng Des 81:827-832
31. Roth J (2005) In: Clark EH, and Reiter DH (eds) Nuclear Fusion Research; Understanding Plasma-Surface Interactions. Springer, Berlin, Heidelberg
32. Grünhagen S, Brennan PD, Knipe S, Stagg R, Yorkshades J, JET-EFDA Contributors (2011) Analysis of hydrocarbons of the JET divertor cryogenic pump at the end of the carbon wall campaign using a micro gas chromatograph. Fusion Sci Technol 60(3):931-936
33. Grünhagen Romanelli S, Brezinsek S, Butler B, Coad JP, Drenik A, Giroud C, 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 CH₄/CD₄ and C₂H_x/C₂D_x at the
398 carbon divertor plates of JT-60U. Nucl Fusion 42:689–696