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Determination of retained tritium from ILW dust particles in JET

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ABSTRACT

Quantitative tritium inventory in dust particles from campaigns in the JET tokamak with the carbon wall (2007–2009) and the ITER-like wall (ILW 2011–2012) were determined by the liquid scintillation counter and the full combustion method. A feature of this full combustion method is that dust particles were covered by a tin (Sn) which reached 2100 K during combustion under oxygen flow. The specific tritium inventory for samples from JET with carbon and with metal walls was measured and found to be similar. However, the total tritium inventory in dust particles from the ILW experiment was significantly smaller in comparison to the carbon wall due to the lower amount of dust particles generated in the presence of metal walls.

1. Introduction

For the preparation of the ITER operational scenarios [1] and the design work of DEMO [2,3], one needs the estimation of tritium retention in deposits on plasma-facing materials (PFMs) and in dust particles generated during the tokamak operation. Beryllium wall in the main chamber and tungsten divertor targets are designed for ITER and metals are candidate materials for the plasma-facing wall in DEMO. However, reports on metal dust particles in present-day fusion devices are limited due to the low number of metal wall machines [4-7]. In particular, analytical data for retained tritium are scarce as they originate from carbon wall machines [8,9]. Analyses for dust particles are difficult in comparison to the study of bulk materials. The micro-size and electrostatic properties of the particles make sampling and handling of the dust challenging. For tritium measurements in dust particles new analytical approaches and techniques are required.

Joint European Torus (JET) started operation with the ITER-like Wall (ILW) in 2011 [10] using tungsten-coated carbon fibre composite (W-CFC) and bulk tungsten divertor tiles, while castellated beryllium blocks and Be-coated Inconel were installed on the main chamber wall. The main aims are: (a) development of an integrated operation scenario with metal walls for ITER; (b) test of material performance with emphasis on power handling, material migration, fuel inventory; (c) development of reactor-oriented engineering solutions especially in the area of fuel cycle and remote handling.

After the first JET-ILW campaign (ILW-1: 2011-2012) dust particles were collected from various places in the vacuum vessel [11,12]. Dust particles in JET contain tritium and beryllium. For analyses of contaminated materials, including dust particles, so-called controlled areas with access restrictions are necessary. These requirements are met by the R&D Building at the Fusion Research Centre (IFERC in Rokkasho) and a JA-EU collaboration [3] could be started in year 2013.

Following the ILW-1 operation, dust particles were collected by means of vacuum cleaning from tiles of 22 divertor modules, i.e. from 92% of the divertor area. In total around 1 g of loose matter was collected: 0.77 g from the inner and 0.27 g from the outer divertor [11]. It should be stressed that these quantities were over two orders of magnitude smaller than the amount generated during the carbon phase

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when 188 g was retrieved from the same area after 2007–2009 operation.

The JET-C and the first JET-ILW campaigns were both performed with the deuterium (D) fueling. However, tritium is present in the machine in low quantities due to the DD reaction and from off-gassing after previous DT campaigns carried out in year 1991 and, especially, in 1997–1998 [13]. As a result, tritium is found on plasma facing components (PFC) and in dust particles. The major aim of this work was the determination and detailed comparison of tritium inventories in dust particles from JET-C and the first JET-ILW campaign. Tritium quantities were determined by liquid scintillation counting (LSC) and using the full combustion method (FCM). The differences between the two experimental campaigns and the impact of location in the divertor on the T level in dust are presented.

2. Experimental

2.1. Dust collection

The dust collections were done during shut-downs using a cyclone type vacuum cleaner. Retrieved dust particles were accumulated in a pot located below a cyclone installed on the vacuum cleaner [14]. In this work, dust particles collected after two different experimental phases in JET, JET-C campaign in 2007–2009 and the first ILW campaign in 2011–2012 have been studied and compared. These dust particles were transported from Culham in UK to Rokkasho Center in Japan in year 2014 [3,15].

Drawings in Fig. 1 show the divertor cross-sections with marked areas of dust collection in the carbon wall phase and ILW-1 in Fig. 1(a) and (b), respectively. In JET-C dust particles were collected from six positions and dust samples from four locations were sent to Rokkasho: inner divertor tiles, carrier ribs, outer divertor base tiles, inner and outer louvres in the remote region of the divertor.

After ILW-1 dust was collected from two areas and respective dust samples were sent to Rokkasho. Dust particles were collected separately from the inner (Tiles HFGC, 1, 3 and 4) and the outer (Tiles 5, 6, 7 and 8) regions in the divertor. In Fig. 1(b) they are marked in red and blue, respectively. Access to remote areas of the divertor was not possible during the shutdown in year 2012. Dust particles at each location were stored in different glass pots. Images in Fig. 2 show the glass pots which were used to transport dust particles. From photographs of the inner divertor dust particles one may infer that these are powders, as shown in Fig. 2(a), whereas in the dust from the outer divertor a mix of flakes (size of a few mm), fluffy and powder material is observed, as shown in Fig. 2(b).

It is stressed that T measurements were done more than seven years after the operation of JET-C and around three years after the retrieval of particles from JET-ILW. Hence, the amounts detected using LSC and FCM are not equal to the amount existing immediately after finishing plasma operations. While the radioactive decay can be taken into account, the effects of T loss by natural off-gassing and possible isotope exchange cannot be estimated. Composition and surface morphology of ILW dust has been reported earlier [12,14] indicating tungsten, carbon and beryllium as the major constituents.

2.2. Sample preparations

Sample preparations were performed in a glove box under a fume hood for both the liquid scintillation counter (LSC) and for the full combustion method (FCM). The work was carried out in a dry nitrogen atmosphere in a glove box. For analyses using liquid scintillation counting a few mg of dust particles were placed into a glass pot which was sealed. Due to limited space in the glove box the mass measurement was done outside the glove box, i.e., in air atmosphere, using a microbalance. As the pre-weight measurement of the glass pot was done in air, the mass difference between the pre-weight measurement and the glass pot containing dry-nitrogen needs to be taken into account. In this case the mass offset is about 1.6 mg. This mass offset was applied to the mass measurements for JET-C particles and ILW dust from the inner divertor.

The amount of dust collected from the JET-ILW outer divertor was very small, as shown in Fig. 2(b), and – because of static charge - it was a difficult task operating in the glove box to collect and transfer particles from the container to the pot. To retrieve dust particles from the glass pot it was necessary to wipe them off using a polycarbonate type membrane filter. This method was applied only for the ILW dust from the outer divertor. In the case of dust collection using the polycarbonate type membrane filter, the mass offset was 2.2 mg. The cocktail solution, Soluene-350, dissolves the polycarbonate type membrane filter, and is therefore not problem for LSC measurement.

Full combustion was performed for particles collected from four locations in JET-C and from two locations after ILW-1 campaign. The samples were placed in 18 small cups made of tin (Sn) foil and wrapped with the same foil in such a way that the dust particles were fully enclosed by the foil.

2.3. Liquid scintillation counter (LSC)

The liquid scintillation counter (LSC) method is a standard technique for quanitification of tritium inventory [16]. In this work, dust particles were put in the cocktail directly and the tritium activity in the ampule was measured using LSC system (Perkin Elmer Corporation). This is a time-effective method used to estimate the tritium amount by measuring β^- radiation (18.59 keV) originating from the surfaces of dust particles. However, the determination of tritium present in the bulk of particles is either not accurate or not possible at all if the studied matter is insoluble in the cocktail.

In general, when particles were in the solution, quenching of signals from LSC system are to be taken into account, as described later in Section 3.2. The standard cocktail of Hionic Fluor and the toluene base solution (Soluene-350) was used to dissolve dust particles and the polycarbonate type membrane filter. Dissolution of the polycarbonate

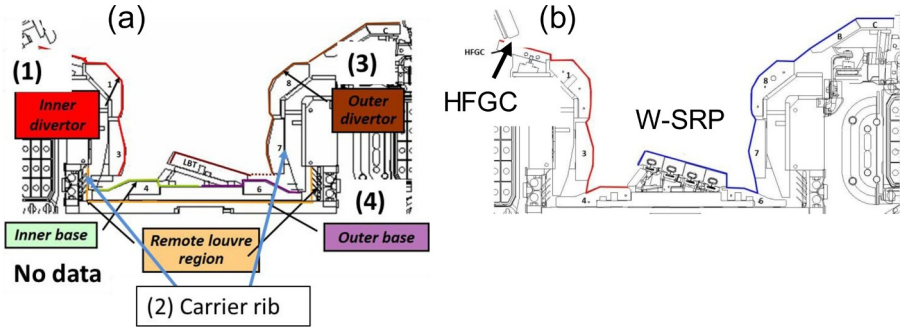

Fig. 1. Schematic drawing of the divertor cross-section with marked areas of dust vacuuming after campaigns in: (a) JET-C 2007–2009 and (b) JET-ILW 2011–2012.

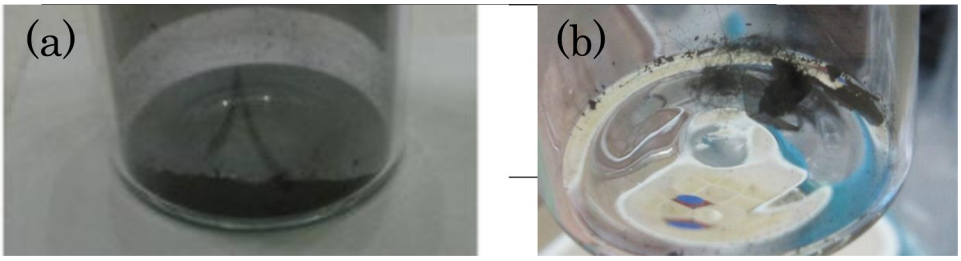

Fig. 2. Glass pots with dust particles from JET-ILW: (a) inner divertor and (b) outer divertor.

type membrane filter was successful, as described in Section 2.2. However, the complex and mixed composition of dust, as described in [12,14], made the complete dissolution of the samples impossible and some solid matter still remained in the cocktails. In general, specimens of a larger mass are better, because a relative error is then decreased, but there is the upper weight limit of 15 mg, as explained in Section 3.2.

2.4. Full combustion method

The full combustion method is also a standard technique for tritium analysis [16]. In general, total retained tritium in bulk materials is determined. In this work, the combustion technique has been used for the first time in studies of dust from fusion devices. The experimental setup used for combusting particles from JET is shown in Fig. 3.

As already told in Section 2.2 dust JET-C and JET-ILW samples were placed in 18 cup-shaped trays made of tin (Sn) foil and the trays were wrapped in such a way that the particles remained inside small packages. Each foil package was placed on a ceramic plate in a quartz tube surrounded by a ceramic furnace. The package was heated up to 1150 K with a heating rate of 85 K/min and kept at a temperature of 1150 K for 30 min. Oxygen flowing through the heated quartz tube was used as a carrier gas for transportation of released tritium to water-filled bubblers.

A feature of the present work was that the tin (Sn) foil was used as a packing of dust particles. It is known that at temperatures of 1100–1200 K tin reacts chemically with oxygen, and during combustion of tin the temperature rises to about 2100 K [17]. Consequently, in our experiments, the Sn foil and dust particles were burned and turned into ash on the ceramic plate.

A water-filled bubbler is used before the heater for humidifying oxygen flowing to the oven. The released tritium was transported as T_2/HT and HTO using an oxygen carrier gas and subsequently was trapped in two water-filled bubblers located downstream and in series of the quartz tube. After switching-off the heating and cooling of the quartz tube, the two downstream bubblers were disconnected and the amount of tritium released from the dust particles during the heating was measured by LSC. In order to measure the amount of tritium remaining in the quartz tube after heating, a third water-filled bubbler was connected downstream of the quartz tube in place of bubblers 1 and 2, and oxygen was passed through the system for 10 min. The third bubbler was disconnected and the amount of tritium remaining in the system was measured by the LSC method. The ash located on the ceramic plate was also put into the cocktail vial to measure the remaining tritium.

2.5. Thermal desorption spectrometry (TDS)

Retention and trapping characteristics of hydrogen isotopes in a small amount of dust particles were evaluated by TDS measurements. The thermal desorption of molecules was measured at a heating rate of 0.5 K/s up to 1273 K by high mass resolution quadrupole mass spectroscopy (QMS) capable of separation between helium and deuterium. Dust particles could not be placed directly in the TDS vacuum chamber. Therefore, a sample of a few milligrams was placed on a tantalum tray with an inner diameter of 8 mm and a depth of 1 mm, which was then inserted into the oven. This method using a tantalum tray for TDS was earlier used for dust particles of JT-60 U [4]. In the system, Mass 4 (D_2) was calibrated using a standard leak of deuterium gas, while other masses were not calibrated. Therefore, the spectrum of Mass 4 was analyzed in this work.

3. Results and discussion

3.1. Quantitative tritium inventories

In the full combustion method a series of water-filled bubblers are used to capture tritium released from the sample by oxidation and trapping of tritiated water. The tritium concentrations measured following the combustion of a 9.5 mg sample of dust retrieved from the louvers in the outer and inner divertor in JET-C was 300×10^3 Bq/cm^3 in the first bubbler connected directly to the oven and 400 Bq/cm^3 in the second bubbler. After removing the first and second bubbler a third bubbler is installed to assess the tritium in the system after heating. The amount of tritium in the system was about 28 Bq. The ash remaining after the combustion was directly inserted into the cocktail. The retained tritium in the ash was 0.6 Bq. This value is twice greater than the

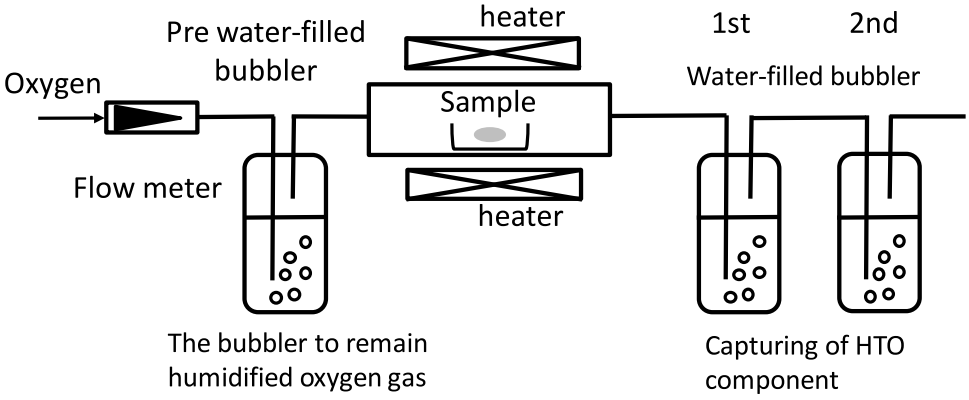

Fig. 3. Experimental set-up for dust combustion. A water-filled bubbler is used also before the heater for humidifying oxygen supplied to the oven. After the heating and combustion of the dust sample is completed, the first and second bubblers are removed for tritium quantification by LSC. A third bubbler is installed and oxygen flowed through. After 10 min the third bubbler is removed for quantification of residual tritium in the system.

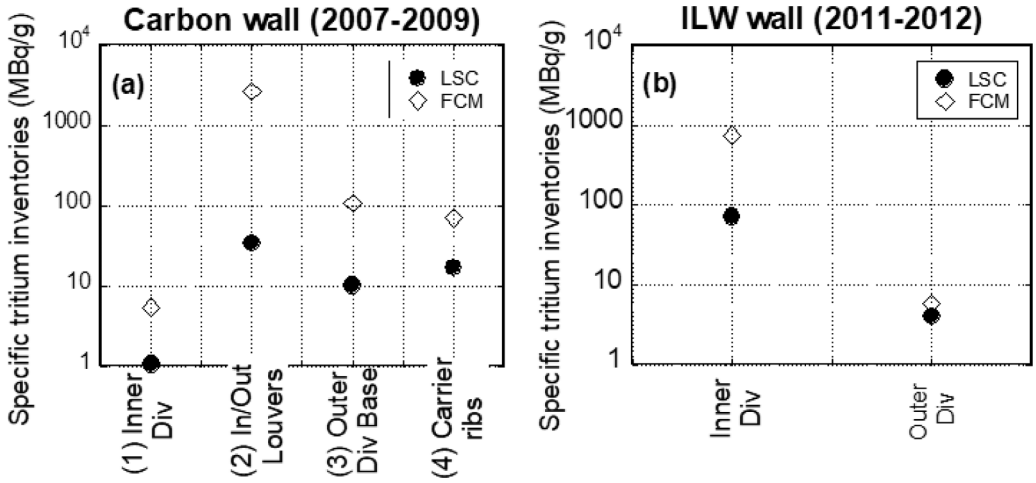

Fig. 4. Specific tritium inventories in dust particles measured by the LSC and the full combustion method (FCM) after campaigns in: (a) JET-C 2007–2009 and (b) JET-ILW 2011–2012 (ILW-1). Note that ILW-1 dust particles at the outer divertor are mixtures containing flakes and powder-type materials, as shown in Fig. 2(b).

T background level (0.3 Bq) determined by oxidizing only tin foil in the same experimental set-up. Therefore, the amount of T retained in the residual ash is negligible. The total tritium was then calculated taking into account results from the first and the second bubbler in that combustion experiment. Using the specific activity of tritium (357×10^{12} Bq/g of tritium) [18], the specific tritium inventory in the dust sample has been assessed at the level of 1.8×10^{18} T atoms/g of dust.

In the case of JET-ILW dust which contains also metallic particles (Be and W) the efficacy of combustion was not clear before starting the experiment. However, with Sn foil used as the heat-enhancing substance in the oxidation, the efficient release of tritium from dust particles could be forced. Graphs in Fig. 4 show specific tritium inventories measured by the LSC and FCM for dust particles from JET-C and JET-ILW, respectively. The comparison of results clearly indicates that the higher tritium amounts are determined by means of full combustion. This comparison also confirms that with the LSC method alone one cannot quantify tritium present in the bulk of dust particles if they are larger than a few micrometers in diameter and insoluble in the cocktails.

The quantities of the collected dust and total T inventories determined by the full combustion method in particles from different locations JET-C and JET-ILW are shown in Fig. 5(a,c) and (b,d), respectively. The mass ratio of dust from those two campaigns exceeds two orders of magnitude with the highest amount retrieved in JET-C from the inner divertor and water-cooled louvers located in the remote areas of the inner and outer divertor. There are large variations in the values of the total tritium inventory (Bq). In JET-C, the greatest is in dust from the louvres (260 GBq) and the lowest in particles from the inner divertor (590 MBq).

As already stated, the amount retrieved from JET-ILW was small, especially from the outer divertor. Total inventories in the ILW-1 dust from two locations are shown Fig. 5(d) with the inner divertor sample having the higher inventory of 580 MBq.

In summary, the specific tritium inventories are in the range 5-2600 MBq/g and 6–750 MBq/g, respectively for JET-C and JET-ILW. The specific inventory for ILW dust was within the same range for JET-C. This relatively high fuel concentration was somewhat unexpected, therefore, the composition of dust and the tritium present in individual particles were investigated further. It has already been shown that matter collected from JET-ILW contained not only W and Be, but also carbon from W-coated CFC divertor tiles and the ribs of the divertor carriers [12,14]. Using the tritium imaging plate technique and electron probe microscopic analysis (EPMA) single dust particles were investigated clearly proving that the presence of T was associated mainly with carbon-based grains: T signal intensity was higher by over two orders of magnitude in carbon than in Be or W particles [19]. This quite reasonably explains why specific activities measured for JET-C and ILW dust samples are similar. Finally, one should stress that the total tritium inventory in the retrieved dust after ILW-1 is only 0.6 GBq, i.e. decreased 450 times with respect to the value of 2.7×10^2 GBq

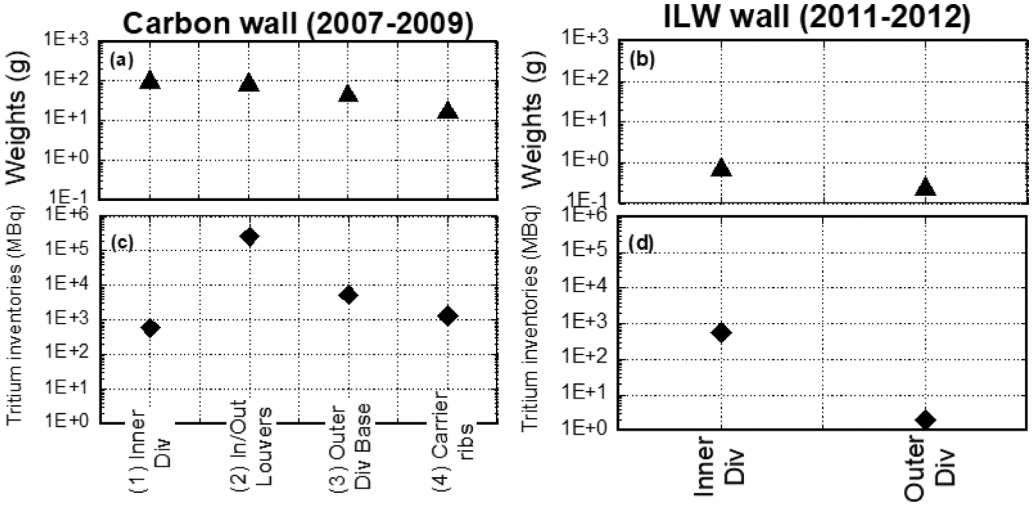

Fig. 5. The mass of dust particles collected at: (a) four locations in JET-C 2007–2009; (b) JET- ILW in 2011–2012. Total tritium inventories obtained by the full combustion method of particles from: (c) JET-C 2007–2009; (d) JET- ILW in 2011–2012 (ILW-1). Note that ILW-1 dust particles at the outer divertor are mixtures containing flakes and powder-type materials, as shown in Fig. 2(b).

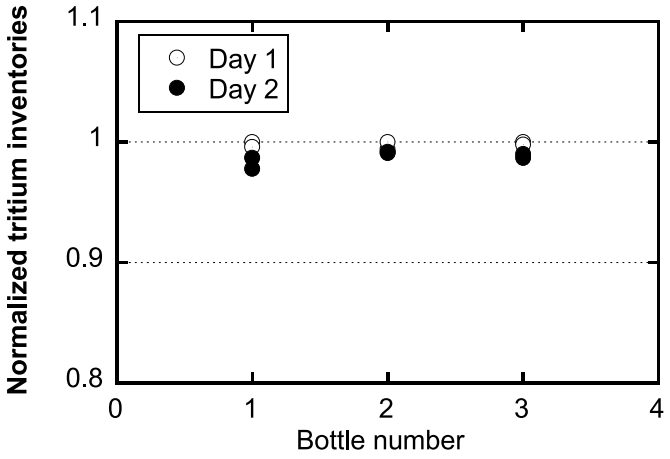

Fig. 6. Normalized tritium inventories in the ILW-1 dust particles measured by the LSC method. Results are for three glass pots with dust particles from the same locations measured four times during two days. The error of the LCS measurements is about 2%.

determined for dust from JET-C. This is a crucial parameter proving very significant differences between dust generation efficiency under operation with metal and carbon walls.

3.2. Sources of errors in tritium measurements

Fig. 6 shows normalized tritium inventories measured by LSC in several samples (few mg each) of the ILW-1 dust from the inner divertor. Four measurements per day were performed during two days. This is the typical time schedule for both LSC and the full combustion method. The standard deviation in T measurements using the LSC is about 2%. In this method the absorption and scattering of β^- radiation in dust particles is of concern. Data in Fig. 6 suggest that in this case the radiation losses did not occur and sufficient signals were obtained. However, when using a larger amount of dust, i.e. more than 15 mg in JET-C dust analyses, signal quenching was observed. The color of cocktails was changed to dark gray or black after shaking the glass pots thus causing color-related quenching and additional scattering of radiation on the particles retained in the liquid. Therefore, when using LCS method for tritium-in-dust analyses the mass of dust must be low (in this case below 15 mg) to prevent significant changes in the color and transparency of the cocktail.

A main source of error arises from the mass measurement using a microbalance. The accuracy of the microbalance is 0.01 mg and the mass variation in the sample preparation is around 0.1 mg. This is related to small changes of conditions in laboratory, especially humidity. For that reason, a mass of more than 1 mg is required for tritium measurements and to ensure a reasonable accuracy of the sample mass.

In summary, one should stress that analyses of T-containing matter must always be performed in a radiation controlled area. This strongly limits flexibility in analytical procedures, the number of qualified laboratories and has a strong impact on the cost, thus making a proper selection of techniques is most crucial. Unfortunately, the literature on T inventory in dust from tokamaks is very limited. Therefore, it was important that the amounts of dust used in both techniques were similar. The accuracy in T quantification by FCM is better than that of LCS. However, taking into account the time required for each analysis method one may state that the LSC method is an effective and relatively simple tool for the estimation of T content in dust particles.

3.3. Thermal desorption

Retention and trapping characteristics of hydrogen isotopes in small amounts of dust particles (4.0–4.4 mg) were evaluated by TDS. The

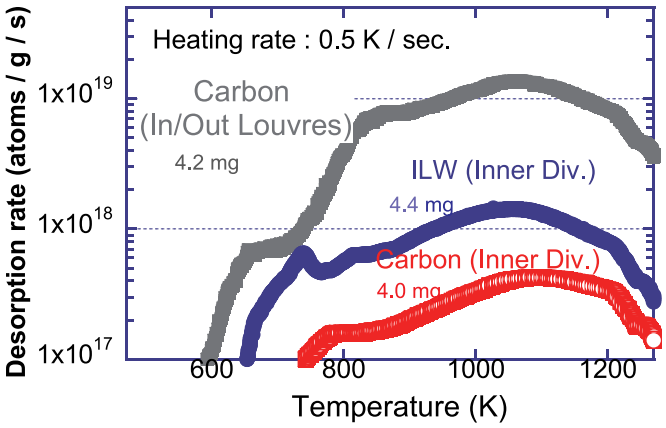

Fig. 7. Thermal desorption spectra of deuterium obtained (from mass 4) for dust particles at two locations in JET-C (2007–2009) and the first ILW campaign (2011–2012).

thermal desorption spectra of Mass 4 (D_2) are shown in Fig. 7. The retention of 1.2×10^{21} atoms/g in ILW-1 inner divertor, 3.7×10^{20} atoms/g in JET-C inner divertor and 1.1×10^{22} atoms/g in dust from the JET-C louvres was measured. D amounts were obtained from Mass 4 in this measurement, due to calibration using a standard leak of Mass 4. Therefore, the D amounts shown here are underestimated because the Mass 3 signal of HD was not taken into account. However, even these reduced data suggest that the amounts of deuterium are 4 to 6 orders of magnitude greater than the tritium concentrations in respective samples.

The maximum release rate is reached in all cases at around 1050 K, but there are also significant differences between the spectra, especially the temperature range for deuterium release. For JET-C desorption starts at 600 K from dust from the louvres but the process begins only at 750 K in the case of matter collected from the inner divertor. It is not possible to attribute these directly to specific species (trapping sites) and any speculation based on a limited number of measurements should be avoided. One may tentatively suggest that particles collected from the inner divertor contained also small graphite grains, i.e. debris from the CFC divertor plates and ribs of the carrier. Such grains would contain deuterium predominantly or even only in the surface region.

4. Conclusion

For the first time ever comprehensive quantitative analyses of tritium have been performed for dust particles collected by a vacuum-cleaner from the JET divertor after operation with carbon (2007–2009) and metal (2011–2012) PFC. The full combustion method allowed for determination of the total and specific T activities in all types of specimens available for the examination. Specific activities were on a similar level for JET-C and JET-ILW because of a significant fraction of carbon in both types of specimens. As revealed by parallel studies using radiography and EPMA, tritium was found predominantly in carbon particles [19]. However, the most important is the drop of the total tritium inventory by a factor of 450 from 2.7×10^2 GBq in JET-C to 0.6 GBq in JET-ILW. This is a consequence of low quantities of dust generated in the operation with metal walls. One should also stress that the full elimination of carbon from a fusion device would most probably result in a further decrease of in-vessel inventory. Another remarkable aspect of this work accomplished at the IFERC-Rokkasho using liquid scintillography and combustion is a provision of grounds in the qualification of analytical methods and procedures for tritium analyses in dust from next-step devices of a reactor class.

Declaration of interests

None

Acknowledgments

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