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Section 13. Hydrogen and helium permeation and release

First wall material issues and related activities at JET

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Abstract

To close the JET tritium material balance a comprehensive programme to determine the tritium inventory in a selected poloidal set of JET tiles was initiated. First results strongly suggest that a significant fraction of the tritium remaining in the machine is immobilised in flakes accumulated in the sub-divertor region. Measurements performed with tiles and flakes retrieved from JET after the DTE1 campaign revealed a chronic tritium release at room temperature, possibly caused by a radiation-induced mechanism. Deposited films of the order of 5 μm thick are found at the vertical surfaces of the inner divertor. Recent analysis of the distribution of beryllium across the divertor shows factors of ≈10 higher beryllium concentrations (relative to carbon) in these deposits than found in the main camber walls. Very little beryllium is found on the outer divertor. Also no beryllium is detectable in the thick deposits accumulated in the shadowed regions of the inner divertor. These results suggest that in ITER migration of beryllium from the main chamber to the divertor surfaces could substantially reduce the chemical erosion and consequently the retention of tritium.

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1. Introduction

Detailed knowledge on the retention and distribution of tritium within the vacuum vessel of tokamaks is important not only for tritium economy reasons but also from the point of view of safety during the operation of large research machines [1]. Thorough information on the fate of tritium inside of machines constitutes the basis for modelling calculations of plasma/wall interactions. Important decisions concerning the selection of the first wall material for ITER largely depend upon the tritium/deuterium experience gained with currently operating machines. Accurate information on the tritium accumulation in various tritium-exposed materials also concerns waste categorisation and disposal issues.

Research activities that may provide solutions to technological open issues related to the first wall materials of JET and ITER are currently underway both at JET and in European national laboratories (Associations). A comprehensive R&D programme covering a wide range of research areas such as tritium in the tokamak, tritium processes and waste management, plasma facing components (PFC), remote handling and inspection techniques, and safety programme was developed under EFDA in close collaboration with the JET Operator (UKAEA) and the Associations.

JET, the only European fusion experiment with practical experience in tritium handling has fuelled far more tritium than any other fusion device worldwide. Tritium studies with first wall materials (FWM) used at JET are therefore particularly valuable. In particular, during the JET deuterium–tritium experiment in JET in

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1997 (known as DTE1 campaign), 35 g tritium entered the torus, of which 6 g remained at the conclusion of the clean-up campaigns [2,3]. A number of experimental programmes profit from this experience. Firstly, to close the JET tritium material balance a comprehensive programme to determine the tritium inventory in a poloidal set of JET tiles was initiated. The results strongly suggest that a significant fraction of the tritium remaining in the machine after the DTE1 campaign is immobilised in flakes accumulated in the sub-divertor region of the machine [4]. Secondly, the tritium distribution, together with studies of deuterium retention and impurity analysis, point to transport mechanisms in the plasma boundary. The implied hydrocarbon chemistry involves reerosion of carbon from the inner divertor wall and migration to shadowed areas [5]. These results suggest that in ITER migration of beryllium from the main chamber to the divertor surfaces could substantially reduce the chemical erosion and the consequent tritium retention. Thirdly, tritiated tiles removed from JET after DTE1 constitute unique materials for studies on detritiation and waste management issues, which will be important acceptance criteria for ITER and subsequent machines.

2. Tritium material balance at JET

Hydrogen isotopes can be trapped in the wall components of fusion devices by essentially three mechanisms:

- Ion implantation in a shallow layer of PFC directly seen by the plasma ions and neutrals. The depth is in the range of several tens of nanometres depending on the material.
- Long range diffusion of hydrogen in the bulk of the PFC, either through the bulk or along grain boundaries.
- Codeposition of hydrogen along with eroded wall material, leading to formation of hydrogen rich deposits. This mechanism depends strongly on the kind of deposited material, its structure and also on the presence of impurities such as oxygen.

With the dominating plasma facing material is graphite such as in JET and various other machines, codeposition has been identified to be the dominant mechanism of long-term hydrogenic retention. Possibly because of this variety of retention mechanisms, not all tritium injected into fusion machines is recovered with the exhaust gas, because a significant fraction is firmly immobilised in tiles, flakes and dust. The trapped fraction in the machine is usually estimated from a comparison between pressure/volume/temperature/concentration (PVT-c) determinations of the input and exhausted gases. They, however, provide only information on the global tritium retention and give no picture of the distribution inside the machine.

In present machines a real-time determination of the hydrogen isotope uptake on in-vessel components during the operation phases of the machine is very difficult or close to impossible. Surface analysis data are mostly available from post-mortem measurements performed during shutdown phases after campaigns have been completed. Results achievable by this procedure give only data on the cumulative uptake after a very large number of pulses. On account of the large inhomogeneity of the distribution of tritium on the surface of tiles, itself strongly dependent upon the position of the tile in the vessel, a very large number of analyses is required for the achievement of dependable results. This is rather challenging because transport and handling of tiles (and even more so the sampling procedure) is associated with the production of fine radioactive dust, requiring the utilisation of glove boxes and concomitant safety procedures.

First wall materials are continually eroded and redeposited by interactions between the plasma and the first wall. As a result some areas of the first wall are net erosion zones, whilst others become covered with deposits that can trap fuelling gases, i.e. the hydrogen isotopes deuterium and tritium, by codeposition. These films may give rise to flakes and dust in the vessel. Most dust is assumed to result from plasma-assisted break-up of deposited films, whilst flakes predominantly result from spalling of progressively accumulating hydrogen/carbon codeposits in colder zones of the machine. The results of a quantitative X-ray microanalysis of flakes collected from JET before the DTE1 campaign is given in [6]. Flakes may have a rather high specific content of tritium (e.g. flakes collected from JET after the DTE1 campaign contained about 1.3 TBq/g [7]). They therefore need to be collected together with the tiles and subjected to a post-mortem analysis to achieve a good tritium material balance. Most redeposition during DTE1 in JET was found to take place on the water-cooled louvres near the inner leg of the divertor. It appears that the majority of the flakes have fallen down to rather inaccessible sub-divertor regions. Their retrieval is difficult requiring specially developed vacuum cleaning devices and remote operation. In a first step preparations are under way for the collection of flakes from one JET octant, where up to 125 g flakes containing of the order of 100 TBq of tritium may be present (allowing for off-gassing losses and radiolytic decay in the years since DTE1). Depending upon the results further efforts will follow in an attempt to recover sufficient tritium for a reclassification of the safety level the machine. According to present knowledge the tritium uptake in structural components inside of the vessel not exposed to the plasma is comparatively small and need not to be contemplated for the tritium balance.

Within the frame of the activities initiated by the EFDA Task Force Fusion Technology a number of reliable techniques for the determination of tritium in samples from tiles and flakes have been developed. After these techniques were tested and verified in practice, an estimation of the total tritium remaining in the machine after DTE1 was carried out. Tiles and specimens from tiles were transported to the properly equipped EU laboratories. Sampling from selected divertor, inner wall, outer wall and limiter tiles was carried out by the so-called 'coring' procedure [8], which essentially consists in cutting cylindrical specimens from the tiles with a hollow drill. To obtain information on the surface (plasma-exposed side) and depth hydrogen isotope content of tiles numerous disks were cut from these cylinders. Disks from the plasma-exposed side of the cylinder yield surface data and all others data from the bulk of the tile. The cylinders as well as the disks were used for the determination of deuterium, tritium, beryllium and carbon. For the analysis of tritium on the plasma-exposed surface and in the bulk of tiles full combustion (FC) and calorimetry (C) were the selected techniques. Ion beam analysis (IBA), secondary ion mass spectroscopy (SIMS) and accelerator mass spectrometry (AMS) were the preferred methods for the analysis of hydrogen isotopes, carbon and metals.

The analysis showed that the plasma-exposed side of all JET divertor tiles contained much less tritium than that contained in the plasma shadowed zones (Fig. 1). This is due to the fact that the plasma-exposed side of the tiles is a zone of net erosion while in the shadowed areas no or negligible erosion takes place.

Good correlation was observed between the results obtained by IBA and FC for deuterium and tritium. Basically, it can be concluded that all techniques provide essentially consistent results concerning the tritium content. Altogether they allow a reliable evaluation of the total tritium content in whole tiles. Both the 'coring' technique and the comparatively high tritium concentrations in tiles from the DTE1 campaign of JET allowed for the first time accurate bulk measurements of FWM previously exposed to deuterium/tritium plasmas. Surprisingly, in some inner divertor tiles, the tritium bulk concentrations were found to be much higher than those on the tile surface FWM [9].

Fig. 1. Tritium content in different locations of the Mark-IIA JET divertor tile number 3 after the DTE1 campaign (including plasma exposed and plasma shadowed sides of the tile).

Some analytical techniques were subjected to a very critical test by careful comparison of numerous tritium measurements performed on a single JET tile. To this effect, several large samples of ≈90 cm^3 were removed from this tile and analysed with a highly sensitive calorimeter (detection limit 0.1 μW). The average specific tritium concentration was $(1.1 \pm 0.2) \times 10^8$ Bq/cm^3. Small cylindrical specimens of about 1.5 cm^3 volume taken from the same tile were fully combusted and the tritium content found to be $(0.7 \pm 0.5) \times 10^8$ Bq/cm^3. Due to the much larger volume of the calorimeter samples, which significantly contributes to average out the non-homogeneous distribution of tritium on the surface, the accuracy of the calorimetry measurements was much higher than that of the FC ones. Irrespective of this, the agreement between the two techniques can be considered satisfactory [10].

Based on the analyses of several hundred specimens, the tritium content remaining in JET tiles after DTE1 was estimated. The tiles with the highest tritium content were found to be divertor tiles 3, 4 and 7 (see Fig. 2). Table 1 gives typical values of the tritium inventories integrated around the torus.

On the basis of measurements such as described above the total tritium remaining in the JET tiles was assessed [4,9]. This inventory (surface and bulk) was

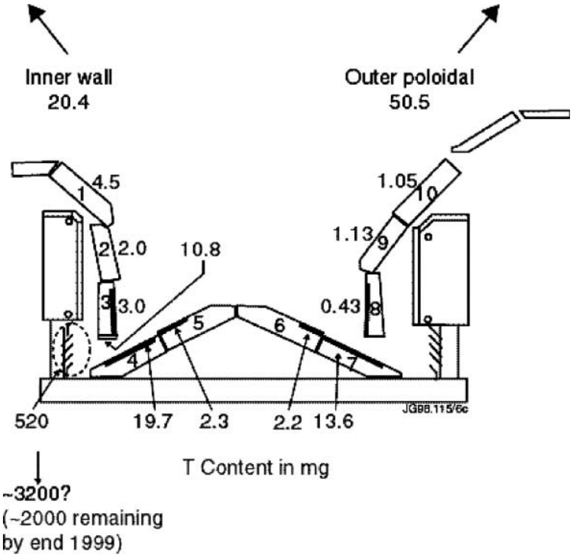

Fig. 2. Tritium content in various regions of the Mark-IIA JET divertor after the DTE1 campaign.

Table 1
Tritium inventories remaining in selected divertor tile toroidal rows after the DTE1 campaign

Divertor tile no. (as shown in Fig. 2)	Total tritium integrated around the torus	
	(g)	(TBq)
3	0.037	13
4	0.053	19
7	0.017	6.2

much lower than originally expected from the overall amount of retained tritium obtained volumetrically. It was thus concluded that more than 90% of the tritium in the machine must be in form of flakes and dust in the sub-divertor region.

Eroded carbon can be deposited in various forms ranging from hard diamond-like a-C:H films to soft polymer-like films which are amorphous, but deposition is also possible in an ordered graphite structure when deposited at high temperature (>2000 °C). The hydrogen uptake of these films depends on the structure and the deposition temperature: hard films contain typically about 0.4 H/C at room temperature decreasing with deposition temperature, whereas soft films can contain H/C ratios well above unity (and decreasing more rapidly with increasing temperature). Tritium is firmly trapped in tiles, flakes and dust requiring temperatures well above 300 °C to achieve a rapid thermal release. Interestingly, a slow chronic liberation of tritium was observed at room temperature, that, in view of the high specific activity of tritium in the flakes, can be attributed to a radiation-induced process [11]. There are also indications for a similar chronic tritium release from tiles.

A comparison between the tritium retention in JET and TFTR was reported recently [12]. At JET material erosion takes place predominantly in the main chamber and flows into the divertor region, where carbon is chemically sputtered or thermally released to migrate into cooler shadowed areas forming thick deposits. On the other hand, in TFTR carbon was eroded from the limiter and the formation of codeposits was also observed. These deposits also spalled off to form flakes, but only years after operations in the machine ceased. The specific activity of the TFTR flakes is much lower than that of JET flakes due to the lower ratio of T:D fuelling over the final TFTR campaigns compared to that in DTE1 at JET.

3. Plasma-surface interactions and erosion

In JET the dominant region of deposition has been at the inner divertor for all divertor configurations (Mark-I, Mark-IIA and Mark-IIGB) [5,13]. Deposition was also at the inner strike-point for X-point configurations prior to installation of the divertor [14]. This behaviour seems to be a unique feature of diverted plasmas as it is observed also in DII, ASDEX and JT-60 [15]. Identification of the net erosion areas is more difficult, but based on probe measurements the entire first wall of JET has been identified as an erosion area [16]. Erosion and deposition in the inner and outer divertor has been modelled with Monte Carlo codes [17,18] based on 'standard assumptions' on the release of C impurities (using physical sputtering plus 2% chemical erosion), impurity transport and redeposition, assuming sticking coefficients of unity of redeposited hydrocarbon fragments and C impurities. These attempts failed to model the erosion/deposition pattern by more than one order of magnitude, predicting deposition near the strike zones and at the side walls with only a slight asymmetry between the inner and outer divertor of less than a factor of two. This result calls for a rigorous reconsidering of the underlying physics assumptions and gives considerable doubt in the confidence of the prediction on erosion/deposition and tritium retention in ITER, which are based on the same assumptions as used for the JET modelling.

Thus the JET data has forced a reexamination of the input parameters for erosion/deposition modelling, and identified three areas where additional physical processes exist that are not correctly modelled. These are drifts in the scrape-off layer (SOL), including plasma wall interaction in the main chamber as an important (or dominant) source of impurities, and a special physics of the hydrocarbon production and transport at the inner divertor [5].

Clear evidence of drift in the SOL has now been observed in JET using the reciprocating probe at the top of the vessel [19], and also in JT-60. The flow, under all diverted plasma conditions, is directed towards the inner divertor, with Mach numbers ranging from 0.5 to 0.7 (Fig. 3). At present there is no final explanation of these drifts but recent modelling suggest $\mathbf{E} \times \mathbf{B}$ drifts in connection with inward directed radial electric fields of about 5 MV/m [20]. Besides probe measurements integrating over long operational periods, erosion of the first wall is manifested in spectroscopic observations of Be (following evaporation onto the wall) in the plasma, and in the divertor (following flux down to the divertor and reerosion from the strike zones).

The carbon and Be impurities eroded in the main chamber are transported by the flows towards the inner divertor where the plasma is normally cold ($T_e < 5$ eV) and dense ($>10^{14}/\text{cm}^3$) leading normally to detached plasmas. Under those conditions soft polymer-like C-films are deposited which exhibit a much stronger chemical erosion as compared with bare graphite or hard a-C:H like films, with erosion yields up to 15%, a value greater those of hard films [21]. In addition to

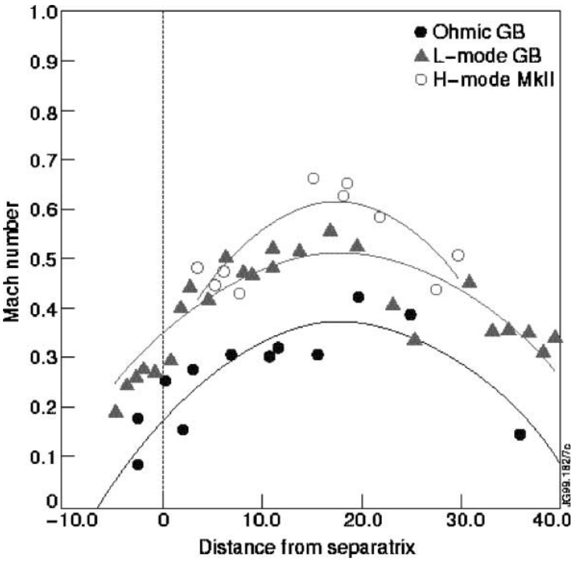

Fig. 3. Drift velocities in the JET SOL under different diverted plasma conditions.

chemical erosion by the background D atoms and ions, these soft films thermally dissolve into a large family of hydrocarbons (mainly C_2 and C_3-type hydrocarbons) for temperatures exceeding about 300 °C. Thus the carbon impurities initially deposited at the inner strike zone (along with the background D ions) are rapidly reeroded. In addition, recent measurements [22] showed very low sticking probabilities of several hydrocarbon fragments when returning back to the surface. Sputtering of the C deposited at the strike zones, together with the low sticking coefficient for some of the hydrocarbons formed, leads to longrange carbon transport in the inner divertor region. This is assisted by reerosion with thermal D-atoms from charge exchange and recombination processes, which impinge more uniformly over the entire surface of the inner divertor. No clear picture exists so far about transport of impurities eroded at the outer divertor. Surface analysis and other data indicate that this is a net erosion area but the amount of redeposited carbon found might not be in line with what we expect to be eroded, indicating a possible transport of carbon out of the outer divertor. However this needs confirmation.

Deposited films of the order of 5 μm thick are found at the vertical surfaces of the inner divertor both for Mark-IIA and Mark-IIGB [5]. Recent analysis of the distribution of beryllium, deuterium and carbon across the divertor shows factors of ≈10 higher beryllium concentrations (relative to carbon) in these deposits than found in the main chamber walls. Very little beryllium is found in the outer divertor. The source of this beryllium is the regular evaporation in the main chamber. The deposition pattern supports the picture of sputtering in the main chamber and entrainment of impurities by strong SOL flows towards the inner divertor. A very important finding of the deposits on the inner vertical tiles is the high Be:C ratio (Fig. 4), when one would expect the incident impurity flux to this area to contain 10 times more carbon. Obviously the beryllium deposited on these areas stays there since it can be transported away only by physical sputtering, which is low or absent at this location due to low plasma temperatures. In contrast, the hydrocarbon chemistry still is active to reerode the impinging carbon impurities, mainly by impact of low energy carbon atoms. Conversely there is no beryllium detectable in the thick deposits found in the shadowed regions of the inner divertor where flaking occurs. In conclusion, since beryllium is not eroded chemically and, once deposited, has a high sticking probability, the migration path of beryllium is restricted to much smaller distances and cannot reach the remote areas.

If ≈50 μm films of carbon are deposited in shadowed areas of the divertor (i.e. 10 times the volume of the 5 μm Be-enhanced films observed), then these will trap significant amounts of the H-isotope fuelling gas. IBA measurements show that D/C ratios of ≈0.8 are typical for such deposits. This would be equivalent to trapping ≈5% of the fuelling gas for the Mark-IIA campaign following which the samples were collected. Estimates of the mean D retention based on pressure and particle balance during the Mark-IIGB campaigns C1–C4 suggest a figure of ≈8% [23]. However, the retention varies with the type of operation, vessel conditions, etc., and may for single days of operation vary from 0% to 20%. It must also be recalled that there is some uncertainty in any one measurement, since it is the difference of quite large numbers. During the DTE1 campaign the T retention was at a rate of ≈40% of the input to the torus [2,3] with no obvious tendency to decrease with time

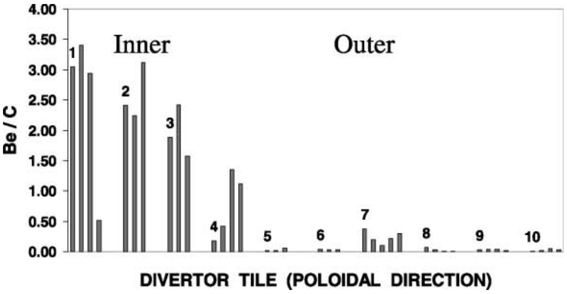

Fig. 4. Concentration ratio Be/C in different positions of the JET Mark-IIA divertor tiles after the DTE1 campaign. The numbers 1–10 in the histogram refer to the tile no. (as shown in Fig. 2), while the four bars within the 10 families of histograms refer each to the poloidal positions in the corresponding tile where the measurement has been obtained.

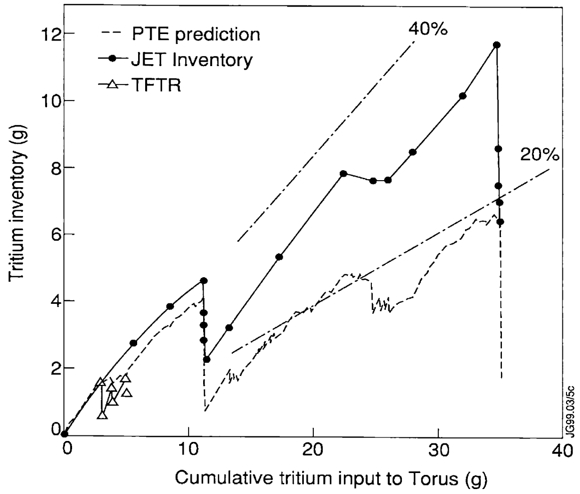

Fig. 5. Tritium balance at JET after the DTE1 campaign. The sharp drop in the curve at about 11 g cumulative tritium input to torus was due to an unpredicted intervention at JET to fix a fault in the neutral beam system.

(Fig. 5). However, the D retention for the same period was typically ≈12% [3]. It is believed that the large T retention is due to isotope exchange of the T with D in the preexisting inventory, which is expected to saturate during continued 50%T/50%D-fuelled operation to give similar rates of T and D retention. This saturation was not observed, although the integrated amount of T fuelling had reached a level comparable to the predicted size of the dynamic inventory. The reason for this is not yet clear.

These results suggest once again that in ITER migration of beryllium from the main chamber to the divertor surfaces could substantially reduce or eliminate the flow of carbon impurities, and thus also the enhanced chemical erosion associated with the deposition of the soft films. This may significantly reduce the consequent tritium retention in ITER. In addition, the Be should also reduce the remaining erosion of the bulk graphite target in ITER. Together, these effects may reduce the transport of carbon to remote areas and the build-up of carbon deposits significantly, but no final answer is so far possible. Furthermore, whether the necessary factor of reduction in T retention in ITER compared to that predicted from JET experiences, which is above 10, can be achieved is not known. To conclusively prove this important point it will be necessary to install a full beryllium first wall at JET.

4. Conclusions

The results obtained at JET strongly suggest that a significant fraction of the tritium remaining in the machine is immobilised in flakes accumulated in the subdivertor region of the machine. In some divertor tiles a significant fraction of tritium is found homogeneously distributed in the bulk. In all cases, tritium immobilised in tiles, flakes or dust is thermally very stable. Under conditions of high specific activity (e.g. flakes) a chronic release possibly due to a radiation-induced process is observed.

The tritium distribution in JET, together with studies of deuterium retention and impurity analysis, point to transport mechanisms in the plasma boundary. Moreover, the implied hydrocarbon chemistry involves reerosion of carbon from the inner divertor wall and migration to shadowed areas. These results suggest that in ITER migration of beryllium from the main chamber to the divertor surfaces could substantially reduce the chemical erosion and the consequent tritium retention in ITER.

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