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Hydrogen isotope inventories in the ASDEX-Upgrade tungsten coated divertor tiles

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Abstract

The total hydrogen isotope inventories in the tungsten coated divertor tiles of ASDEX-Upgrade were measured by thermal desorption spectroscopy (TDS) and – in the near surface region – by nuclear reaction analysis (NRA). The highest amounts of deuterium ($\leqslant 5 \times 10^{22}$ D/m^2) are measured in the inner divertor region outside the separatrix due to the formation of a C:H layer on the tungsten by codeposition of deuterium with carbon. The inventories in the outer divertor – where erosion dominates – are generally smaller by an order of magnitude. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Apart from graphite and beryllium, the high-Z material tungsten is considered as a plasma facing material for ITER. In order to provide an experimental database for the ITER divertor, tungsten coated graphite tiles were installed as divertor target plates in the ASDEX-Upgrade tokamak for the experimental period December 1995–July 1996 [1].

In order to obtain a measure and to improve the understanding of hydrogen retention in tungsten under reactor-like conditions, the total hydrogen isotope inventories of the tungsten divertor tiles of ASDEX-Upgrade were measured with thermal desorption spectroscopy (TDS). The near-surface deuterium inventory was measured by nuclear reaction analysis (NRA).

2. Experiment

The tungsten coated carbon tiles had been installed in the ASDEX-Upgrade divertor at the separatrix intersection areas (Fig. 1). They consist of a 500 μm thick plasma sprayed tungsten layer on a graphite substrate. In order to avoid hot spots at the edges, the tiles were tilted in toroidal direction. Hence, a part of each tile is shadowed from the plasma by the edge of the adjacent tile, and receives much less plasma flux than the non-shadowed part of the tile. In the following, we will denote the former region as 'shadow region', whereas the region with the full plasma impact will be denoted as 'plasma region'.

About 800 discharges were performed in the tungsten experimental period. Out of these, about 480 discharges were heated additionally by neutral injection. Initially, the heat load on the tungsten tiles was limited to 7 MW/m^2 in order to avoid damage, but during the experiments, discharges with the full auxiliary heating power of 10 MW were also performed, resulting in heat loads of more than 10 MW/m^2 [1].

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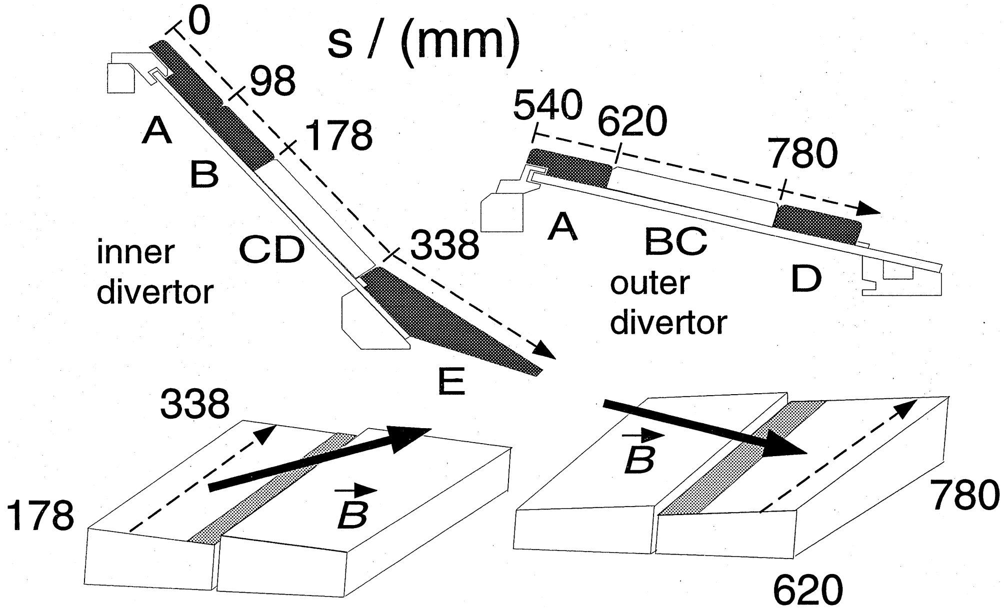

Fig. 1. Position of the tungsten coated graphite tiles (CD and BC, respectively) in the ASDEX Upgrade divertor. The tiles are tilted in toroidal direction, resulting in regions shadowed from the plasma by the edge of the adjacent tile. The arrows denote the directions of the magnetic field lines. The outer graphite tiles had no tungsten coating.

Mostly, the discharges had deuterium as the filling gas, and, in case of neutral injection, as the injected atoms as well. Hydrogen as filling gas was used mainly in the middle of the experimental period and for the last 10 discharges. In the case of deuterium filling, hydrogen (H) is still present in the discharge. Typical values are 10–20%, while in the case of neutral injection with H^0, the H background amounts to about 60% [2]. From these figures and the number of different discharges, a mean H/D ratio in the sum of all discharges in the plasma core of about 0.5 can be estimated.

2.1. Thermal desorption spectroscopy

The experimental setup used in this work for the TDS experiments is described in Ref. [3]. The samples, having a diameter of about 12 mm and a thickness of about 3 mm, are cut out of the tungsten coated tiles and are heated by electron bombardment from the rear side up to 2100 K in an ultra high vacuum chamber. The desorbed gases are monitored by means of a calibrated quadrupole mass spectrometer (QMS) by the increase of the respective partial pressures, at corresponding masses, during the heating of the sample. The total hydrogen isotope inventories are obtained by adding up the respective atoms in all of the released hydrogen containing molecules (e.g. HD, D_2 and CD_4 for deuterium).

Table 1 shows the results of laboratory TDS experiments of hydrogen implanted in graphite or tungsten including peak temperatures T_{peak}, bonding types, binding energies E, and trap concentrations [4–6]. TDS spectra of hydrogen implanted in graphite show a broad peak in the range of 900–1200 K, indicating the strong C–H bonds with energies of 2–4 eV [4]. TDS spectra of hydrogen desorbed from tungsten after implantation at room temperature reveal a two peak structure [5–8]. The explanation of this peak structure is still under discussion. A first peak at about 500–600 K may be attributed to intrinsic traps [5] with concentrations of the order of $\leqslant 0.01$ traps/W-atom and binding energies of about 0.5 eV [5]. The second peak at about 700–800 K occurs only after implantation with energies sufficient to produce damage (see below); hence, this peak may be due to trapping at the damage sites with concentrations in the range of 0.1–0.2 traps/W-atom and trapping energies of about 1.5 eV [5]. During implantation – in contrast to graphite – the solute amount of hydrogen in tungsten is of the same order as the trapped one; this solute amount is released after the implantation [9] and can be seen in TDS spectra by an immediate rise of the released flux after the onset of the temperature ramp, when TDS is started shortly after the end of implantation. In the case of the ASDEX Upgrade samples, we can expect, that the solute amount of hydrogen in tungsten was totally released during the storage of the samples (several months). The hydrogen isotope ions impinging the divertor plates have energies of the order of 100 eV, resulting in a mean range of less than 10 nm [5]. Hence, damage in the tungsten divertor tiles is not produced by the impinging deuterium ions, but due to the impact of

Table 1
Laboratory TDS results of hydrogen implanted in graphite/tungsten with information about peak temperature T_{peak}, bonding types, binding energies E, and trap concentration [4–6]

	T_{peak} (K)	Bonding type	E (eV)	Concentration (traps/host-atom)
Graphite	900–1200	C–H	2–4	0.4 (implantation zone) $\leqslant 0.01$ (intrinsic, at crystalline surface)
Tungsten	500–600	W, intrinsic	0.5	$\leqslant 0.01$
	700–800	W, ion induced	1.5	0.1–0.2

multiple charged impurity ions (mainly carbon) from the plasma [1]. However, surface damage in the tungsten due to the impact of deuterium ions with energies of some 100 eV is possible by other mechanisms as formations of bubbles [10] and displacement of intrinsic impurities [11].

2.2. Surface-layer analysis

Thick deposited carbon containing layers on the tungsten surface were detected by means of 2 MeV H^+ Rutherford backscattering for all samples from the inner divertor. The thickness of these layers was estimated to be several μm, without large poloidal variations. In contrast, erosion dominates in the outer divertor: these samples hardly show any carbon deposition at all. The actual surface composition (mainly boron, carbon, oxygen, and tungsten) was determined by X-ray photoelectron spectroscopy (XPS). The largest amounts of boron (40%) and oxygen (20%) were detected in the plasma region of the inner divertor. However, even for the erosion dominated outer divertor samples, boron and oxygen are still present at the very surface [12].

Before performing TDS, the near-surface deuterium inventory was measured by NRA using the 790 keV $^3He(d,\alpha)p$ reaction, resulting in an analyzing range of 0.5 μm (for pure tungsten) to 2 μm (for an amorphous co-deposited hydrocarbon layer). Hence, the analysis depth depends on the not well-known surface composition of the respective sample, leading to a relatively high uncertainty of the NRA results. Due to the larger surface roughness of the plasma-sprayed tungsten coatings, no deuterium depth profiles can be obtained. If deuterium containing layers with thicknesses exceeding the analyzing range are involved, NRA can only establish a lower limit for the results to be expected from TDS.

3. Results and discussion

As in the case of the samples from the previously installed graphite divertor [2], the release of hydrogen isotopes from the tungsten coated tiles during TDS is dominated by H. The amounts are in the range of about (5–8) × 10^{22} H/m^2 for all samples. The by far largest part of this H can be attributed to the water uptake both on the tungsten front surface as well as on the graphite back surface during the storage of the samples in air [2]. Hence, we will restrict the following discussion on deuterium.

3.1. Deuterium inventories

Fig. 2 shows the deuterium inventories for the different regions of the inner and outer divertor as measured by TDS and NRA. Also shown are the distributions of the mean surface temperature during the discharges with auxiliary heating by neutral injection and the incoming fluence of deuterium, as measured by thermography [13] and Langmuir probes [14], respectively. The latter distributions show corresponding maxima at about 310 and 725 mm, roughly indicating the mean position of the separatrix during the tungsten experimental period.

The highest deuterium inventories (about 5×10^{22} D/m^2) are measured in the shadowed region of the inner divertor outside the separatrix. At the position of the separatrix, the inventory shows a minimum, although the amount of deposited carbon (as measured by Rutherford backscattering) shows only minor poloidal variations. The inventories of the outer divertor samples are generally smaller than the inner divertor inventories by a factor of 10; here the total inventories, measured by TDS, indicate a maximum at the separatrix position, whereas the inventories in the near surface region, measured by NRA, show a minimum at the separatrix. Having in mind the results of the surface analysis we can conclude, that deuterium retention in the case of the ASDEX-Upgrade tungsten divertor tiles in the inner divertor is governed by co-deposition of deuterium with carbon ions from the plasma, whereas implantation into the tungsten bulk governs the retention in the outer divertor. Co-deposition was also found outside the separatrix in the graphite tiles of the inner divertor [2]. The co-deposited layers of the inner divertor tiles are depleted of hydrogen at the separatrix position as well as in the plasma region due to the increased temperature. Furthermore, the deuterium inventories in the co-deposited layers are of the same order of magnitude as those in the graphite divertor tiles ($\leqslant 2 \times 10^{23}$ D/m^2) [2],

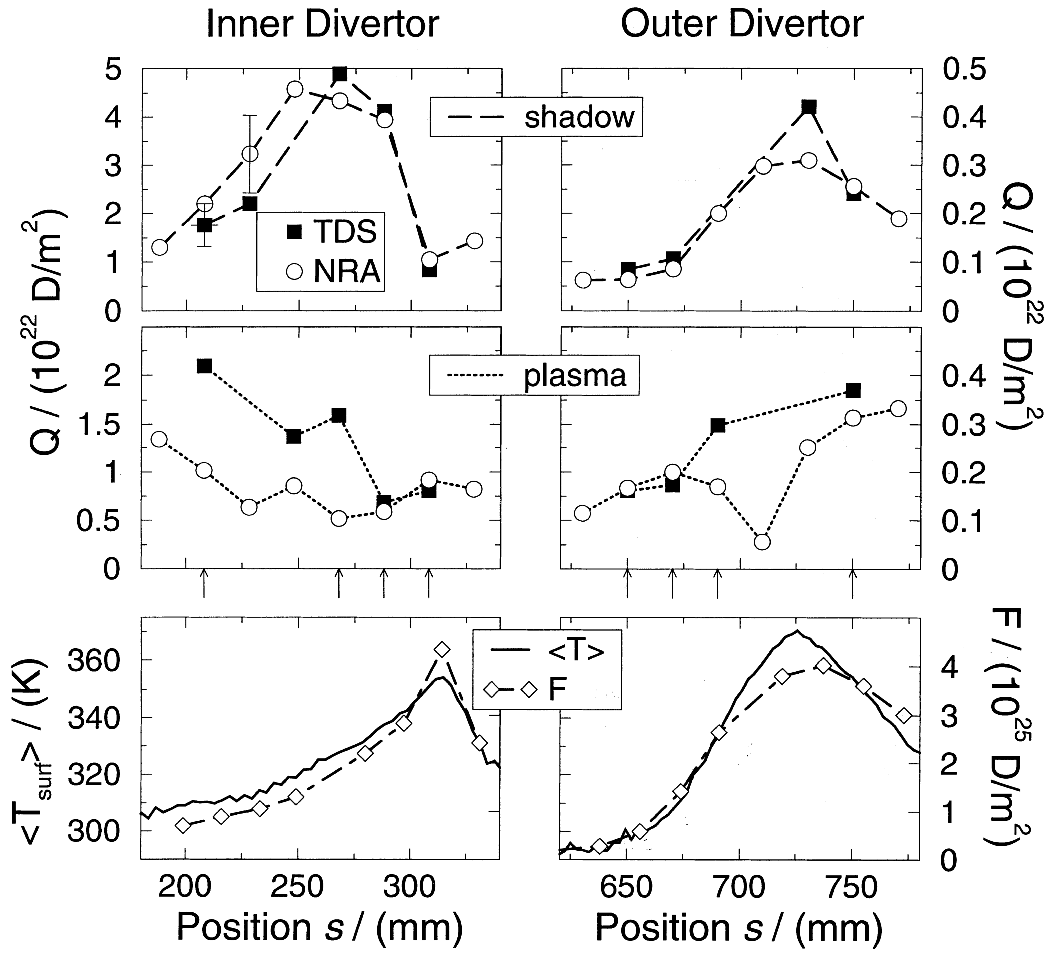

Fig. 2. Distribution of the total D inventories (Q), measured by NRA and TDS, at the shadowed and the plasma regions of the tungsten coated tiles, respectively. Also shown are the distributions of mean surface temperature ($\langle T_{surf} \rangle$), and the total incident D fluence (F). Note the different scales for the deuterium inventory plots.

taking into account the shorter exposure time of the tungsten coated tiles (about 700 compared to about 2000 discharges).

The total inventory in the tungsten coated divertor samples differs from the near surface inventory by only a factor of less than 2, except for the separatrix region of the outer divertor and the plasma region of the inner divertor outside the separatrix. This is in contrast to the graphite divertor samples, where the total inventory exceeds the near surface inventory by a factor of up to 100 indicating diffusion of large amounts of hydrogen out of the co-deposited/implanted layer into the graphite bulk to depths of some 100 μm [2]. The differences between the results of TDS and NRA can be explained by the thickness of the co-deposited layer being larger than the analysis depth of NRA. The fact, that the total inventories in the separatrix region of the outer divertor exceed the near-surface inventories, and that the TDS spectra reveal deuterium retained in the underlying graphite substrate only for the outer plasma separatrix region (see below), indicates that deuterium diffuses into the bulk of tungsten with ranges of some few μm (hence exceeding the implantation range by more than a factor of 1000), as it is assumed in models describing the behavior of hydrogen in tungsten [5]. The combination of high fluxes and high temperatures at the outer separatrix region depletes the near surface region, but also results in a large amount of deuterium which can diffuse into the tungsten bulk and in the underlying graphite substrate (see below).

3.2. TDS spectra of deuterium

Fig. 3 shows some examples of TDS spectra of the tungsten coated divertor samples. For the different regions in the inner and outer divertor, (at least) four

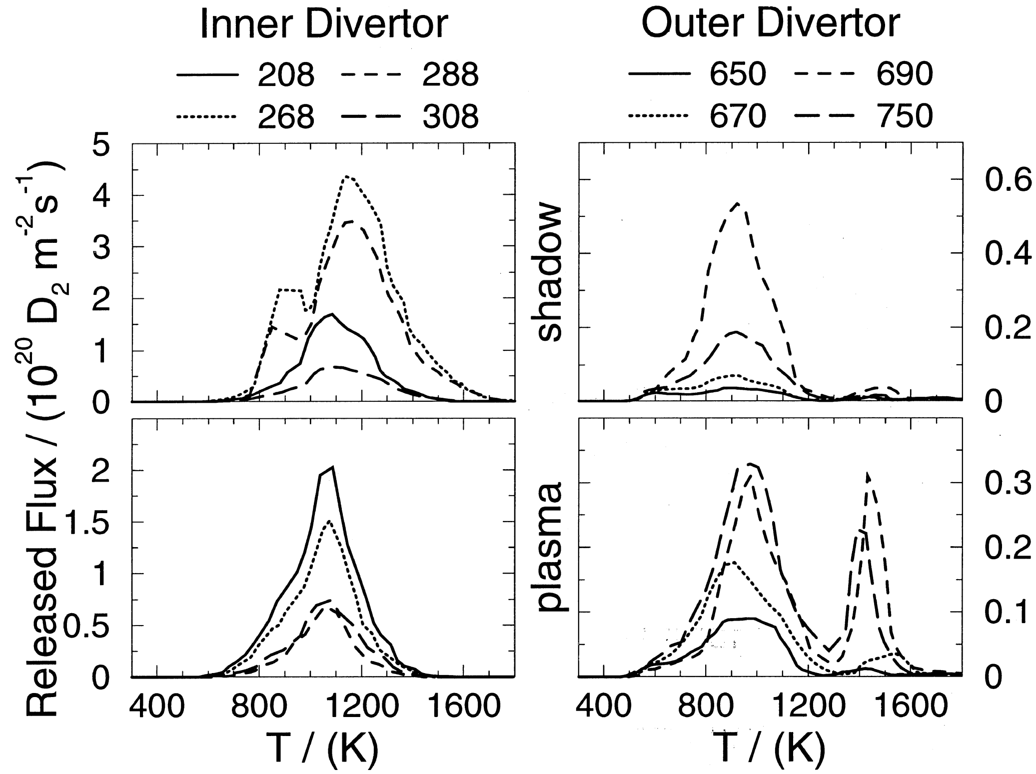

Fig. 3. Released fluxes of D_2 during the heating of the samples for the shadowed and plasma regions of inner and outer divertor tiles. Heating ramps are of the order of 10 K/s. The legend numbers denote the sample positions, as indicated by the arrows in Fig. 2.

different peaks can be distinguished and attributed to different deuterium retention mechanisms (Table 1): (1) a small peak (shoulder) at about 600 K, as in the case of the outer divertor samples, due to trapping at intrinsic trapping sites in the tungsten layer; (2) a broader peak at about 900 K in the case of the samples from the outer divertor, due to trapping at ion induced damage sites in the tungsten layer; (3) a broad and large peak at 1000–1100 K in the case of the samples from the inner divertor, due to C–H bonds in the co-deposited a-C:H layer – in agreement with the peak temperature of the D_2 release from the graphite divertor samples [2]; and (4) a sharp peak at about 1400 K in the case of the samples from the outer divertor, which might be due to trapping of deuterium in the underlying graphite substrate after diffusion through the tungsten layer. The latter retention mechanism is pronounced in the case of the samples from the separatrix region ($s = 690$, 748 mm), where due to high temperatures and fluxes a relatively large amount of deuterium can diffuse through the tungsten layer. Generally, the peak positions are shifted to higher temperatures compared to laboratory results, indicating the diffusion of some deuterium out of deep regions of the samples.

Fig. 4 shows the relative amount of deuterium in the different trapping sites of the tungsten divertor tiles obtained by Gaussian deconvolution of the TDS spectra. For the inner divertor more than 90% of the deuterium is trapped in co-deposited C–H layers except for the plasma region near the separatrix. In the plasma region of the inner divertor near the separatrix up to 40% of the deuterium is trapped at ion induced damage sites in tungsten. For the outer divertor the main part of the deuterium (up to 90%) is trapped at ion induced damage sites in tungsten. Only at the separatrix position of the plasma region about 40% of the deuterium is trapped in the underlying graphite substrate. Furthermore there is some trapping at intrinsic carbon, introduced by the manufacturing process, in tungsten.

3.3. Balance of deuterium

In order to have an estimate for the tritium inventory in ITER we calculated the retained amount of deuterium in the divertor tiles and first wall of ASDEX-Upgrade. Table 2 shows the input of deuterium into ASDEX-Upgrade through the gas valves and by neutral beam injection (NBI) and the retained amount of deuterium

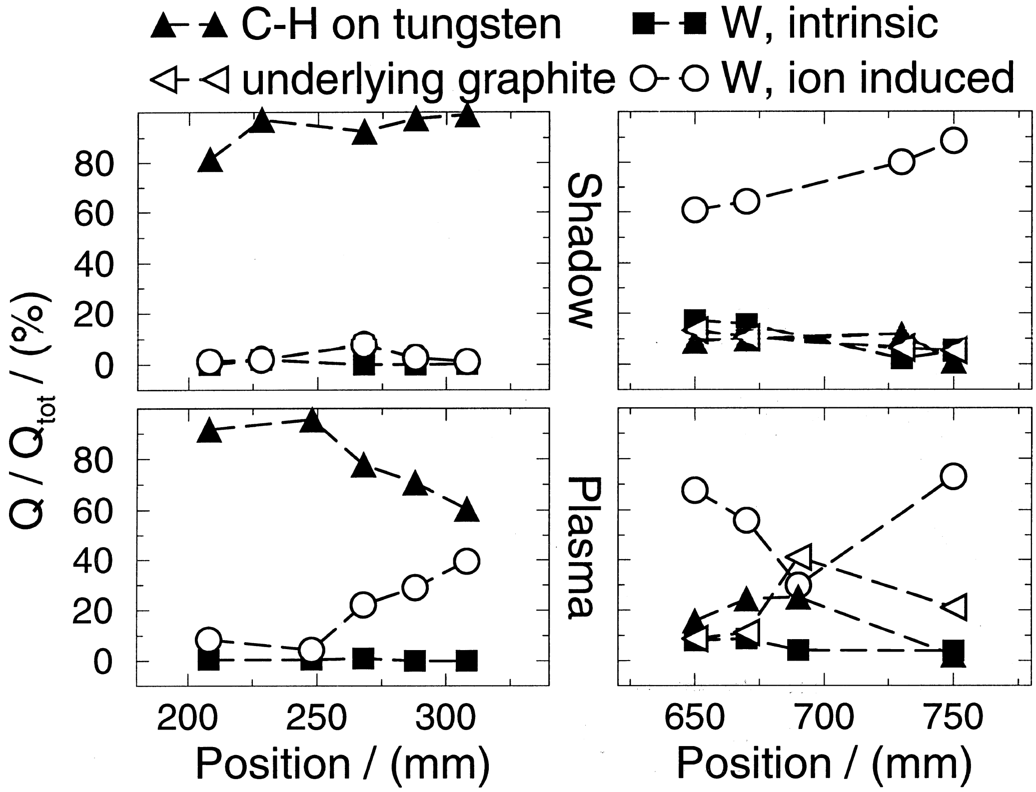

Fig. 4. Relative amount of deuterium in the different trapping sites of the tungsten divertor tiles (Table 1) as obtained by Gaussian deconvolution of the TDS spectra.

during the tungsten experimental period. With these numbers, about 2% of the total deuterium input was retained in the tiles of the inner wall of ASDEX-Upgrade including more than 1% retained in the tungsten divertor tiles [15]. This is in contrast to measurements at TFTR and DIII-D where about 40% of the deuterium input can be found in the first wall [16].

Table 2
Gas balance of deuterium in ASDEX-Upgrade during the tungsten experimental period. Mainly deuterium was puffed through the gas valves. A minor amount was injected by the neutral beams (NBI). About 2% of the input has found in the first wall of ASDEX-Upgrade

Input [15]		
Gas values	7.1×10^{24} D	
NBI	0.6×10^{24} D	
$\sum$	7.7×10^{24} D	
Retained amounts of input		
W divertor tiles	1.0×10^{23} D	1.3%
Other graphite tiles [15]	0.5×10^{23} D	0.7%
$\sum$ first wall	1.5×10^{23} D	2.0%

4. Summary

The total hydrogen isotope inventories of the tungsten coated divertor tiles of ASDEX-Upgrade – consisting of a 500 μm thick plasma sprayed tungsten layer on a graphite substrate – were measured with quantitative TDS. The near surface deuterium inventory (≈1 μm) was measured by NRA using the 790 keV ^{3}He(d,α)p reaction.

The total hydrogen isotope inventory measured 'in' the tungsten divertor tiles of ASDEX-Upgrade is dominated in the case of the inner divertor by the build-up of thick co-deposited C–H layers (several μm) onto the tungsten surface outside the separatrix. The inventories in these co-deposited layers are of the same order of magnitude as the total hydrogen inventory measured in the previously installed graphite tiles. Increased temperatures, as in the separatrix region, deplete the co-deposited layer, reducing the deuterium inventory by a factor of 5. Diffusion of hydrogen out of the co-deposited C–H layer into the underlying tungsten layer is hindered by the tungsten/carbon interface. The inventories in the outer divertor samples are generally smaller by a factor of 10 than the inner divertor inventories; here the retention is governed by implantation and diffusion out of the implantation zone into the tungsten bulk. In the case of high fluences and high temperatures (separatrix), some hydrogen seems to diffuse into the graphite substrate.

Co-deposition with carbon from the plasma is the by far most effective hydrogen isotope retention mechanism for the tungsten coated divertor tiles installed in ASDEX-Upgrade. This also will be the case in future fusion devices, as long as carbon is the main plasma impurity.

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