Hydrogen Retention in Tungsten Materials Studied by Laser Induced Desorption*

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Abstract:

Development of methods to characterise the first wall in ITER and future fusion devices without removal of wall tiles is important to support safety assessments for tritium retention and dust production and to understand plasma wall processes in general. Laser based techniques are presently under investigation to provide these requirements, among which Laser Induced Desorption Spectroscopy (LIDS) is proposed to measure the deuterium and tritium load of the plasma facing surfaces by thermal desorption and spectroscopic detection of the desorbed fuel in the edge of the fusion plasma. The method relies on its capability to desorb the hydrogen isotopes in a laser heated spot. The application of LID on bulk tungsten targets exposed to a wide range of deuterium fluxes, fluences and impact energies under different surface temperatures is investigated in this paper. The results are compared with Thermal Desorption Spectrometry (TDS), Nuclear Reaction Analysis (NRA) and a diffusion model.

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1. Introduction:

Tritium retention in plasma facing components (PFCs) due to plasma wall interactions is one of the most critical safety issues for ITER and future fusion devices. For carbon based PFCs the co-deposition of fuel with re-deposited carbon has been identified as the main retention mechanism. This retention grows linearly with particle fluence and can reach such large amounts that carbon is most probably omitted in the activated phase of ITER and future reactors. Instead, tungsten is foreseen as PFC material in the divertor of ITER and is the most promising candidate of PFCs in future reactors. Its fuel retention behaviour is subject of present R&D.

In this work the retention of fuel (deuterium) in bulk tungsten and the ability to desorb the retained fuel by millisecond spot laser desorption (LID) has been studied. The W materials were exposed in several plasma devices, such as in the edge of the TEXTOR tokamak [1], the linear plasma generator Pilot-PSI [2] and small scale plasma devices, covering several orders of magnitude in ion flux and different range of temperatures. Some of the comparisons were done by keeping the fluence similar (ca. 10^{25} D/m^2), achieved at different fluxes and thus different exposure times, concentrating on flux dependence. Laser Induced Desorption up to temperatures of typically 2000 K has been used as main analysis method. This technique achieves good spatial resolution (3-5 mm) without sample cutting, but with an analysis depth of typically up to 100 μm. For comparison, laser heating was also performed up to surface melting, showing in some cases additional fuel release, as well as conventional TDS and NRA.

2. Analysis methods

In situ detection of retained fuel by LIDS relies on the thermal desorption of the retained fuel

by laser heating and its detection by spectroscopy in the plasma. To crosscheck the methods, the same laser pulse has been directed on the same samples either in a laboratory device or in the tokamak TEXTOR. Most results are obtained by LID where a small spot is heated up to 1800-2000 K in 1-3 ms. The released hydrogen isotopes are monitored in laboratory experiments by a residual gas analyzer (RGA) [3] and in TEXTOR by Balmer lines spectroscopy (LIDS) [4] of the injected particles (H, D) in the edge plasma. Both methods are calibrated absolutely allowing the determination of the retained H and D particles (atoms and molecules).

For both applications, a Nd:YAG laser with variable pulse duration and up to 20 kW power at maximum 60 J energy per pulse has been used . Additionally, the repetition rate is limited by the maximum average power of 300 W. The laser light is coupled into fibres of 400 μm diameter either to the laboratory experiment or to the tokamak. The fibre cross-section is imaged onto the sample surface producing a nearly top-hat distribution of the power density up to 500 MW/m^2 with a temporal decay of 30% over 3 ms. The laser properties presented in Fig. 1 are obtained in laboratory with an imaging lens of 6 cm focal length with a distance of 43.3 cm (for 2 mm spot diameter) and 56.8 cm (for 3 mm spot diameter) between fibre end and target surface.

To optimise the accuracy of the mass spectroscopic detection, the vacuum chamber is decoupled from the pumping system by closing the gate valve shortly before the laser pulse and the partial pressure increase of all desorbed gas molecules is measured by a residual gas analyzer (RGA) scanning from mass 1 to 50 within 2.5 s. The RGA signals are calibrated against a calibrated leak with H_2, D_2, CH_4, CD_4 etc. in the proper range.

3. Hydrogen retention and laser desorption in tungsten

To apply LIDS for in situ monitoring of tritium retention, it is of fundamental interest to

qualify and understand the desorption process during the laser spot heating. Previous LID experiments have been performed on bulk graphite and hydrocarbon layers up to a thickness of 15 μm showing that more than 90% of trapped hydrogen is released in a single laser shot. This is consistent with the model of local thermal detrapping of fuel from binding states with local recombination and rapid outward transport as molecule [5]. However, retention and release of fuel in bulk W is based on implantation, long range diffusion and trapping and possible release by recombination of atoms on the W surface [6]. Model calculations of hydrogen diffusion and trapping during laser induced heat pulses on tungsten using the one dimensional code TMAP7 [7] show a strong dependence of the amount of desorbed particles on the initial fuel penetration and the assumptions on the binding energy. The fuel penetration depends strongly on the exposure temperature, weakly on the fluence, but not on the energy of the injected particles. Assuming an initial deuterium profile of 9 μm depth with 3 μm decay length, which is observed by NRA depth profiling by other groups on similarly loaded samples, exposed to a standard laser heat pulse of 500 MW/m^2 for 3 ms, about 86% of the deuterium inventory is desorbed in a single laser pulse. The simulation was done with a diffusion coefficient of $4.1 \cdot 10^{-7} \cdot \exp(-0.39\ eV/k/T)$ according to [8], which models only the interstitial traps with 0.39 eV binding energy. Assuming additionally traps of 1% of lattice density with 1.45 eV binding energy in the tungsten bulk reduces the hydrogen release from 86% to 53% in a single laser pulse (Fig. 2).

To investigate this behaviour experimentally, different tungsten targets were loaded with deuterium at different temperatures and fluences. All tungsten samples have been treated before exposure to 1273 K in vacuum for more than one hour to remove the initial hydrogen isotopes and anneal material stresses. Additionally only deuterium was selected for retention in tungsten to distinguish it from environmental protium.

4. Results and discussion

In the TEXTOR tokamak, a tungsten target of 4 cm x 6 cm (unpolished, polycrystalline, rolled tungsten with 99.95% purity provided by Goodfellow GmbH) was fixed on a roof limiter with the surface normal tilted 36^{o} with respect to the toroidal B-field. The limiter was transferred into the main plasma chamber from the bottom and the tip was positioned 1.3 cm behind the last closed magnetic surface (LCMS). The target was exposed to a maximal fluence of $1.7 \cdot 10^{25}$ D/m^2 at the tip surface within 110 s plasma with a decay of fluence along the target in the scrape off layer (SOL) [9]. The radial flux dependence at the target is presented in Fig. 4. During plasma exposure maximum temperature excursions from 400 K to 700 K were observed at the tip. A radial temperature gradient is established during exposure, while the temperature is constant in poloidal direction. The electron temperature of 30 eV at the limiter tip results in deuterium impact energies of up to 140-180 eV due to sheath acceleration.

The tungsten plate was removed from the vacuum chamber and inserted into the laboratory device for LID measurements. Fig. 3 shows a picture of the tungsten plate after LID measurement. A spot of 3 mm diameter was heated up to 1800 K by a laser pulse with 10 J absorbed energy in 3 ms corresponding to 500 MW/m^2 (standard LID pulse). To assess fuel which possibly was not released in these pulses, the central area of the spot was molten by reducing the spot size to 2 mm diameter and increasing the energy to 15 J corresponding to 1.6 GW/m^2. Previous experiments have verified that all retained D in the molten material is released.

Two different series were performed. In row (a) the tungsten was molten where all D inventory in the exposed area is removed. In row (b) the deuterium was desorbed up to

temperatures of 1800 K first by a standard laser pulse followed by a second laser pulse with high power density which led to laser induced melting (LIM) in the spot centre. The results of the measurements are presented in Fig. 4, showing that D signals normalized to the exposed surface area in row (b) are always lower than the signal from the molten spots in row (a). This already indicates that laser desorption is not complete and pushes some amount of deuterium out of the volume accessible for desorption. Generally the inventory decreases with increasing plasma radius and decreasing particle fluence, as expected. However, the retention increases again at 48.4 cm. This is correlated with the start of the deposition of a ca. 30-50 nm thin carbon layer at larger plasma radii, visible also in Fig. 3. The carbon layer retains larger amount of hydrogen when exposure time and fluence increase, but acts also as a protection barrier for deuterium diffusing into the W material.

The ratio of the amount desorbed until 1800 K to the total amount of D is plotted in Fig. 4. In the erosion dominated area, this value increases from 25% to 42% and jumps up to about 90% in the deposition dominated area. The increase can be explained by a decreasing surface temperature and/or particle fluence. This would be consistent with the model calculations where higher temperatures increase the diffusion depth with the consequence that a smaller fraction of D is desorbed by the LID pulse. In the deposition zone the majority of D is retained in the carbon layer acting as a barrier for further penetration of D atoms into the W bulk. Here the high release fraction in LID is very similar to the results obtained in a-C:D layers deposited on carbon material [10].

The possible influence of the target temperature on D retention in tungsten and its desorption by LID has been further investigated at other samples. In the Pilot-PSI facility [2] a tungsten target (polycrystalline, rolled and polished tungsten, 99.95% purity provided by Goodfellow GmbH) was exposed to fluxes up to $2 \cdot 10^{24}$ /m^{2}s over 10 s with an energy of 1.2 eV

additionally accelerated by a negative biasing voltage of 55 V. These fluence and particle energy are comparable with those in the erosion zone of the TEXTOR sample. Despite the high heat load the surface temperature was controlled by water cooling reaching 655 K in the centre and 620 K at the edge.

Flux measurements were performed in vertical direction by Thomson scattering whereas an LID profile of retained D was obtained in horizontal direction. The data indicate a small deviation between the plasma beam and target centre, which is also observed by the IR camera measurements. The radial distribution of the fluence and the desorbed deuterium are presented in Fig. 5. With the standard laser pulse a maximum value of $6 \cdot 10^{19}/m^2$ deuterium atoms was desorbed in the first pulse. This is nearly identical with the value observed in the erosion zone of the TEXTOR sample, applying the standard laser heating pulse.

Fig. 5 shows in addition the profile of desorbed deuterium in a second laser pulse on the same spots, which was always below 10% of the amount in the first pulse. In a third series part of the same spot was molten to release D which may not have been released in these laser pulses. The ratio of the desorbed deuterium in the first laser exposure to the total amount of stored deuterium is given in Fig. 5. It drops from 85% at the edge down to 63% in the target centre. This is consistent with the results in the TEXTOR experiment indicating an inverse dependence of the desorbed particles on the target temperature. The increase of the flux density by a factor of 10 does obviously not compensate the increased desorption fraction induced by the lower exposure temperature.

An unpolished tungsten target (polycrystalline, hot-rolled tungsten with 99.97% purity provided by Plansee GmbH) was exposed in an ECR deuterium discharge [11] with 38 eV/D and a flux of $1 \cdot 10^{20}/ m^2s$ for 18 hours reaching a fluence of $6 \cdot 10^{24}/m^2$. The temperature was held constant to 370 K during exposure. LID measurements were performed with standard laser parameters showing a D retention of $6.8 \cdot 10^{20}$ D/m^2 in the first pulse as shown in Fig. 6.

With consecutive laser pulses on the same spot, the LID signals decays from 10% in the second pulse to about 1% after 10 laser pulses. The additional release and slow decrease of these values is attributed to lateral and perpendicular D diffusion into the desorption volume. LID at other locations at the same target delivered nearly the same results. In order to determine the fraction of desorbed D atoms by the 1st pulse of LID measurements in relation to the total amount, not only laser induced melting, but also TDS and NRA were applied. In TDS measurements [12] with heating rates of 0.3-600 K/min up to 1273 K, an integrated amount of desorbed deuterium of $7.5 \cdot 10^{20}/m^2$ was found, corresponding to 91% of the deuterium measured by LID within the 1st laser exposure.

In addition, the D concentration was measured with NRA [13] at two energies (6 MeV ^{3}He with 12 μm penetration depth and 2 MeV ^{3}He with 7 μm penetration depth) and the D profile reconstructed by the code NRADC [14], delivering $5.5 \cdot 10^{20}$ D/m^2 and $4.1 \cdot 10^{20}$D/m^2, respectively. Inside the laser spot about $0.7 \cdot 10^{20}$ D/m^2 was measured in the first 7 μm. This corresponds to about 17% of the deuterium retention in the sample outside the spot (Fig. 7). These results are in fair agreement with the results by LID and TDS. Inside the laser spot, the NRA depth distribution shows a strong depletion of D by a factor of 1000 at the surfaces as predicted by the model calculations (Fig. 2).

The high fraction of released deuterium by LID in this case indicates again the increase of the released fraction with decreasing temperature, but might be also supported by the low flux in this case.

To further elucidate this, two unpolished W targets (polycrystalline, rolled W, 99.95% purity provided by Goodfellow GmbH) were exposed to deuterium in a RF D_2 glow discharge with a flux of $2.8 \cdot 10^{19}/m^2s$ for 2 hours reaching a fluence of $2 \cdot 10^{23}/m^2$. The sample was biased with 500 V simulating D impact energies similar as in the TEXTOR SOL plasma (150-250 eV) and the temperature was held constant at 533 K during exposure. LID measurements showed

$4.7 \cdot 10^{19}$ D/m^2 in the first laser pulse and $0.5 \cdot 10^{19}$ D/m^2 in the second pulse. TDS measurement results in $7.35 \cdot 10^{19}$ D/m^2, so that 65% of retained D was detected by LID. Although the fluence in this target was lower than in the ECR exposed sample, LID was not able to desorb all D atoms. The major parameter again was the 163 K higher target temperature during deuterium loading supporting further the assumption that the temperature is the main parameter which determines the penetration and population of traps and by this the fraction of release accessible for LID. As a consequence less deuterium can be desorbed by LID compared to LIM and TDS.

5. Summary

Tungsten samples have been exposed under different conditions (cf. Table 1) to deuterium plasmas and the amount of retention was measured by Laser Induced Desorption (LID) with mass spectroscopic detection and compared with conventional results from TDS and NRA. Laser heating has been performed in standard pulses up to about 2000 K in 3 ms and also up to melting of the laser spot.

For deuterium fluences between $(2\text{-}200) \cdot 10^{23}$ D/m^2 the absolute retention in bulk W extends between $(0.74\text{-}7.5) \cdot 10^{20}$D/m^2, in reasonable agreement with the data from literature [15]. For W samples exposed at temperatures < 400 K, a good agreement is found between NRA, TDS and LID. With increasing surface temperature during exposure, TDS, NRA and LIM found systematically larger amounts of deuterium retention compared with LID under standard conditions (heating up to 1800 K). The results can be explained by the fact that with higher surface temperature and exposure time the deuterium diffuses deeper into the W bulk and/or is stored in traps with higher binding energy. For both cases some of the retained deuterium is not accessible anymore for the desorption by the standard LID laser pulse heating. This behaviour is in line with results from the TMAP modelling. This effect must be

considered for the application of this method for W-bulk retention on erosion dominated areas. It has been found that Laser Induced Melting desorbs all the retained D (under the above conditions) and can be an alternative for complete desorption of hydrogen from W. In deposition dominated areas the developing a-C:H layer can act as a barrier for retention of fuel in W.

Apart from the exposure temperature, flux and fluence, additional parameters might influence the retention, e.g. the surface topography, microstructure of the W bulk material and the plasma impurities.

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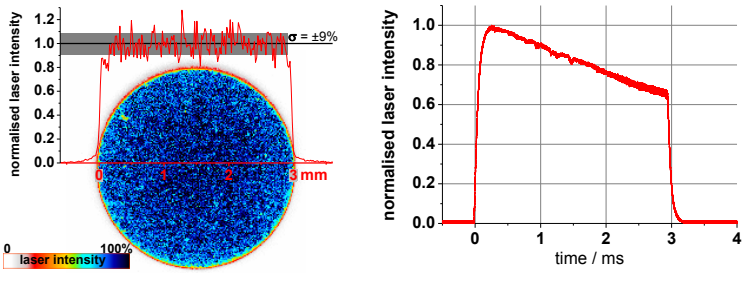

Fig. 1: (a) Spatial distribution and (b) temporal development of the Nd:YAG laser intensity after transmission through a 35 m long fibre of 400 µm core diameter (optical magnification 7.5 times)

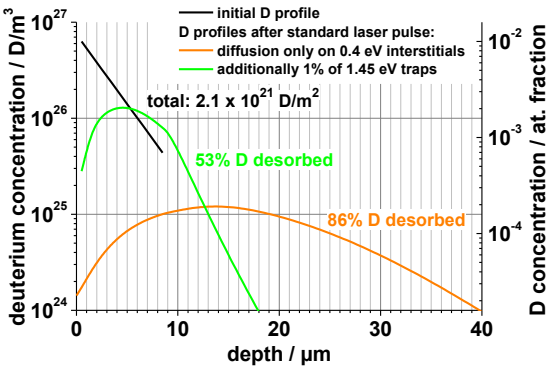

Fig. 2: Model calculation for the hydrogen concentration in tungsten bulk after laser irradiation considering traps with different binding energy

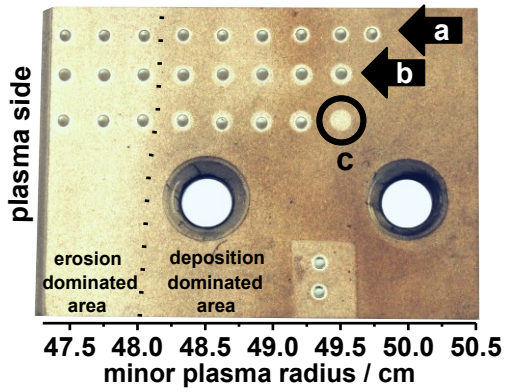

Fig. 3: W target exposed in TEXTOR. Row a: laser induced melting (LIM), row b: 1st pulse desorption (LID) and 2nd pulse melting (LIM), c: desorption only (LID)

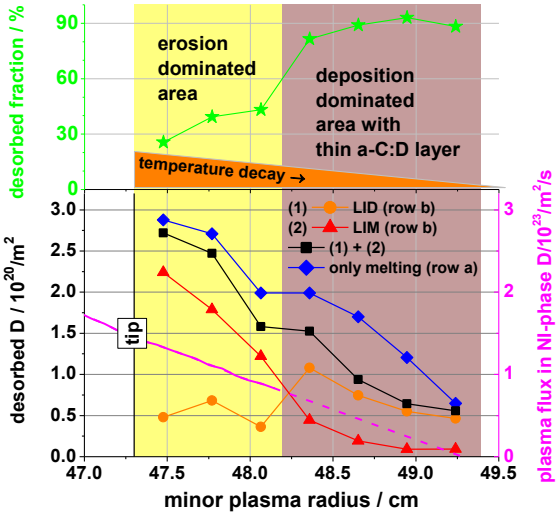

Fig. 4: Left axis: LID of TEXTOR W sample (RGA results) and desorption efficiency; right axis: flux dependence during exposure

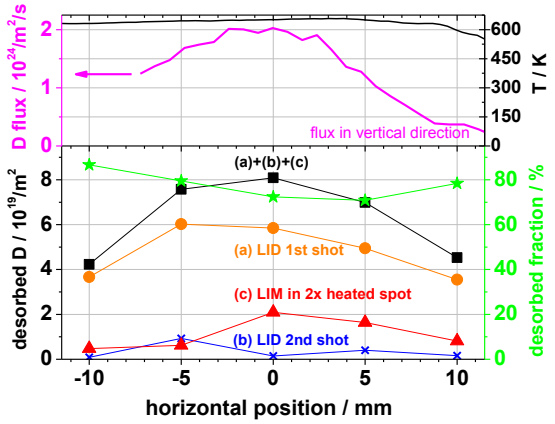

Fig. 5: Radial profile of exposure conditions and LID results on a tungsten target exposed in Pilot-PSI

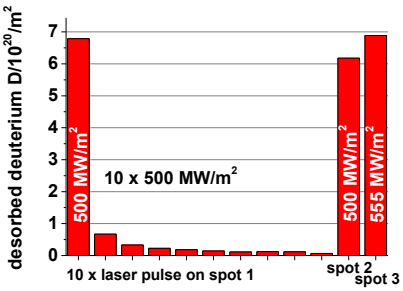

Fig. 6: Effect of lateral and perpendicular diffusion in successive laser pulses

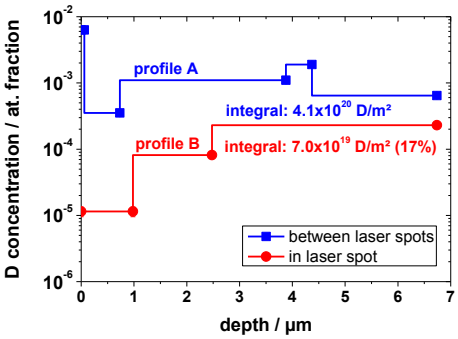

Fig. 7: NRA depth profiles of D concentration in a W target exposed in an ECR discharge with long exposure time at low temperature

plasma type ► and device	tokamak (TEXTOR)	linear device (Pilot-PSI)	ECR plasma	RF glow discharge
exposure:				
flux / D/m^2/s	$1.5 \cdot 10^{23}$	$2 \cdot 10^{24}$	10^{20}	$2.8 \cdot 10^{19}$
fluence / D/m^2	$1.7 \cdot 10^{25}$	$2 \cdot 10^{25}$	$6 \cdot 10^{24}$	$2 \cdot 10^{23}$
exposure time	110 s	10 s	64800 s	7200 s
temperature	400-700 K	620-655 K	370 K	533 K
ion energy	60-180 eV	55 eV	38 eV	350 eV
desorption:				
D inventory / D/m^2	$0.5\text{-}2.7 \cdot 10^{20}$ (LIM)	$4\text{-}8 \cdot 10^{19}$ (LIM)	$7.5 \cdot 10^{20}$ (TDS)	$7.35 \cdot 10^{19}$ (TDS)
D fraction de-sorbed by LID	25% - 93%	63% - 85%	91%	65%

Table 1: Overview of exposure conditions and retention results in the order of appearance in the text