Available online at www.sciencedirect.com
ScienceDirect
Journal of Nuclear Materials 363–365 (2007) 1236–1240
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Deuterium retention and surface modification of tungsten macrobrush samples exposed in FTU Tokamak

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Abstract

The effect of discrete structures such as macrobrush or castellated surfaces on power handling and deuterium retention of plasma facing components is to be assessed since such geometrical configurations are needed for increasing the lifetime of the armour to heat-sink joint. Four small macrobrush W and W + 1%La_2O_3 samples have been exposed in the Frascati Tokamak Upgrade (FTU) scrape-off layer up to the last closed flux surface by means of the Sample Introduction System. FTU is an all metal machine with no carbon source inside vacuum vessel; it exhibits ITER relevant energy and particle fluxes on the plasma facing components. Here, results on morphological surface changes (SEM), chemical composition (EDX) and deuterium retention (TDS) are reported.

PACS: 52.55.−s; 52.40.Hf; 68.43.−h; 68.43.Vx

Keywords: Deuterium retention; FTU; Material probe; Tungsten; Castellation

1. Introduction

Macrobrush structures or castellated surfaces are presently being considered for plasma facing components to be installed in the ITER divertor and baffle regions. Stress release and reduction of eddy currents are advantageous features of this solution. On the other hand, some concern exists about impurity and fuel retention inside gaps of neighbouring elements as well as enhanced interaction with plasma because of the sharp edges, arc initiation from the corners, formation of bridges across the gaps in case of melting [1].

To the purpose of studying morphological changes and deuterium retention induced by plasma operation, four small macrobrush W and W + 1%La_2O_3 samples have been exposed in the FTU scrape-off layer up to the last closed flux surface, by means of the Sample Introduction System [2]. FTU is an all metal machine with no carbon source inside vacuum vessel; it exhibits ITER relevant energy and particle fluxes on the plasma facing components.

2. Experimental layout

Four samples, with size $15 \times 15 \times 37\ mm^3$, two made of pure W and two made of W + 1%La_2O_3, were provided by EFDA. They were shaped in form

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0022-3115/$ - see front matter
doi:10.1016/j.jnucmat.2007.01.256

of a macrobrush, with four 'teeth', 10 mm long. Unfortunately, the narrowness of access to the FTU vacuum vessel ruled out larger sample dimensions.

As received samples were analysed by SEM and EDX. Before analysis all the samples were baked for an hour at 1173 K, under vacuum. Higher baking temperature was not used for avoiding recrystallization of tungsten. The presence of a large quantity of C and O was detected by EDX measurements. The ratio between W and C corresponds to their ratio in tungsten subcarbide (W_2C), which is known to be formed during annealing under UHV conditions above 1070 K [3]. The large content of C and O in all the samples, much higher than expected in a W bar produced from powder metallurgy, cannot be only due to the surface contamination by air exposure: EDX analysis of overheated regions in samples exposed to air contamination after removal from FTU, shows much lower or null content of O. Furthermore, for one of the sample, EDX analysis with half of the electron energy was carried out resulting in negligible difference as compared to the full energy, thus confirming that oxygen and carbon are homogeneously distributed, at least in the first half micron.

Because of the crowded FTU experimental programme and the relatively high number of discharges needed for a detectable amount of retained deuterium, sample exposure in FTU was mainly allowed in a parasitic way. Therefore, all the samples were exposed also to discharges with unintentional strong plasma interaction and to disruptions. In particular strong heating and surface melting of the first two samples occurred, resulting in a hardly valuable loss of retained deuterium. As a consequence, TDS data as well as ion fluence values are reported only for sample 3 and 4. Even for these samples, deuterium inventory is to be considered a lower limit. Sample 3 was exposed to 46 discharges (8 disruptive), sample 4 to 55 discharges (12 disruptive).

The samples were exposed in the FTU scrape-off layer, at major radius $R = R_0$ and with the leading edge at minor radius between $r = 300$ mm and $r = 310$ mm (the radius of the toroidal limiter is 290 mm). Due to the slight elongation of the plasma in FTU, the sample tip, for $r = 300$ mm, was almost tangent to the last closed flux surface (LCFS), defined by the inboard toroidal limiter. No sample temperature measurement could be implemented.

The main plasma parameters were: plasma density $n_e = 0.5\text{–}1.0 \times 10^{20}\ m^{-3}$, plasma current $I_p = 0.35\text{–}0.5$ MA, toroidal magnetic field $B_T = 4\text{–}6$ Tesla. Most of the discharges were run with $n_e = 0.5 \times 10^{20}\ m^{-3}$, $I_p = 0.5$ MA, and $B_T = 6$ T. By assuming a typical FTU particle confinement time $\tau_p = 3 \times 10^{-2}$ s, an approximate evaluation of the total deuterium ion flux Γ_D to the samples can be performed according to:

$$\Gamma_D = (n_e * V) * (A_{sample}/A_{TL})/\tau_p \ (\text{ion s}^{-1}), \qquad (1)$$

where V is the plasma volume, A_{sample} and A_{TL} the sample and the toroidal limiter cross section area, respectively.

By summing the contribution of each discharge, we get, for sample 3 and 4, 3.5×10^{20} and 6.1×10^{20} impinging deuterium ions, respectively.

3. Results and discussion

In Fig. 1, the photo of sample 1, taken after the exposure in FTU, is visible. Every sample shows traces of melting: in particular, on sample 1 and 2 huge melting is visible on the top of the teeth. Nevertheless, the gaps are still open, like before exposure, without 'bridges' between the teeth: in someway castellation seems to hold melt in place. No arc track is recognised on the samples. Sometimes non deep cracks and/or small holes (submicron size) are seen on the surface.

In the molten and nearby zones, the usual recrystallization of the material is visible with grain enlargement and highlighted grain boundaries. At the boundary of molten zones deep cracks are visible, probably as a result of tensile stresses during resolidification of melted material. In not melted,

Fig. 1. Sample 1 after exposure in the FTU scrape-off layer (the sample is shown inserted in the sample carrier).

but however overheated zones close to the sample tip, a pattern of nearly spherical, sometimes agglomerate together, tungsten granules, of submicron size is recognizable (see Fig. 2). Similar tungsten granules have been found on a solid tungsten plate exposed to the TEXTOR plasma [4]. In [5,6] microscopic droplets are said to be left by rapid solidification of melted material during disruption tests.

From post-exposure EDX analysis, the main results are:

(1) in the zones of the samples far from the top, on both sides facing ion and electron side, and in not overheated or melted part of the top, the near surface content of W, C, O is rather similar to the one before exposure;
(2) in melted (or close to molten material) zones oxygen and lanthanum are absent and the concentration of W is 100% or much larger than the one before exposure, carbon being the only element detected besides W;
(3) the percent composition of the elements, in a zone of sample shadowed from the plasma, is identical to the one before exposure.

It should be taken into account that each sample was exposed to many steady state discharges after the latter of the suffered disruptions (which are probably the main responsible for melting and overheating); therefore the post-exposure absence of oxygen in melted, or close to melted, zones, suggests that negligible contribution to the oxygen content of the samples is to be associated with impurity ion fluxes in the FTU scrape-off layer. Carbon exhibits a large variety of concentrations, probably related to the persistence or not of tungsten carbides: in any case, no carbon source exists in FTU vacuum vessel.

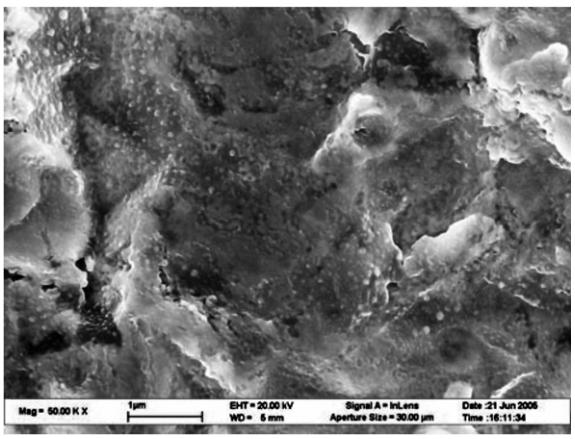

Fig. 2. SEM image of sample 3 – upper, non melted zone.

The amount of retained deuterium was measured by thermal desorption spectroscopy. The possible influence of castellation on deuterium retention was looked for by trying to compare our results with literature data about not castellated samples.

The samples were heated up to 1173 K by a ramp rate of 6 K/min. The amount of D retained in the sample was determined by integrating the quadrupole mass spectrometer signal for D_2, HD and CD_4 during thermal desorption. The CD_4 signal was monitored because of the large presence of C in the samples: however in both samples its contribution to the total number of desorbed deuterium atoms was limited.

In Figs. 3(a) and 4, the flow rate of D_2, HD and CD_4 is reported for sample 4 and 3, respectively, in terms of molecules/s as a function of the elapsed time, together with the sample temperature. A different behavior of D_2 and HD spectrum between sample 3 and 4 can be easily recognized. Sample 3 exhibits only a broad desorption peak at about 800 K for both D_2 and HD. On the other hand, sample 4 shows two, even though not well resolved, peaks at about 470 and 850 K, with decreasing amplitude, for both D_2 and HD. A third peak, at about 600 K, is only visible for HD, while a very shallow peak at about 1170 K is also present for D_2. Desorption of HD molecules, as found in [7], increases with temperature also beyond the monitoring time (total amount of retained deuterium is therefore slightly underestimated). The rather higher time constant of HD release, as compared to the D_2 release, suggests that the HD molecules are probably formed at the walls of the TDS chamber by recombination of D outgoing from the sample with H adsorbed at the walls.

The temperature location of the release peaks depends on implantation temperature, particle energy, ion flux and heating rate: in literature, a large range of temperatures is reported [8]. As a consequence of the temperature dependent dynamic desorption during implantation, samples implanted at lower temperature usually show desorption peaks at lower temperature with respect to samples implanted at higher temperature. Unfortunately, as above said, no sample temperature measurement could be performed during exposure in FTU. No large difference in heat loading should be expected

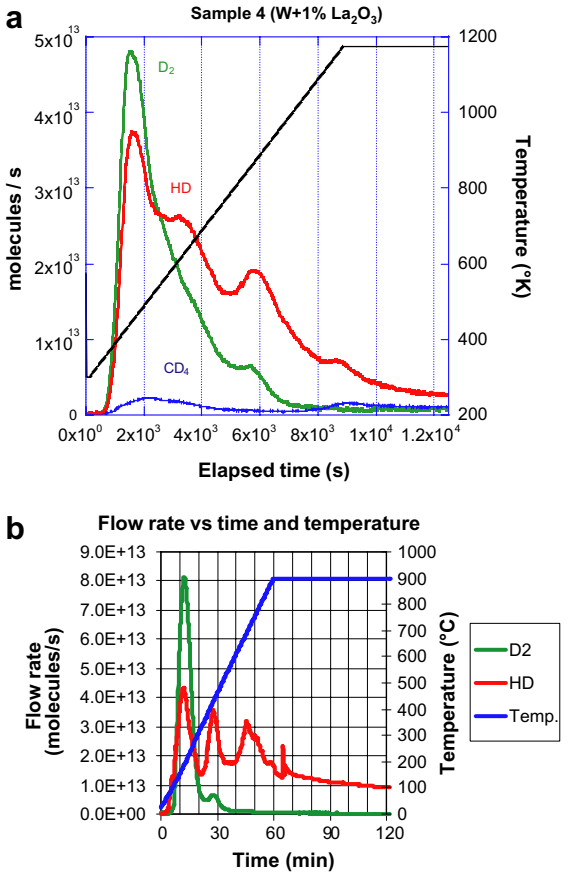

Fig. 3. Flow rate (molecules/s) of D_2, HD and CD_4 as a function of the elapsed time, together with the sample temperature, for (a) sample 4 of present work and (b) sample implanted at 773 K (from Ref. [9]).

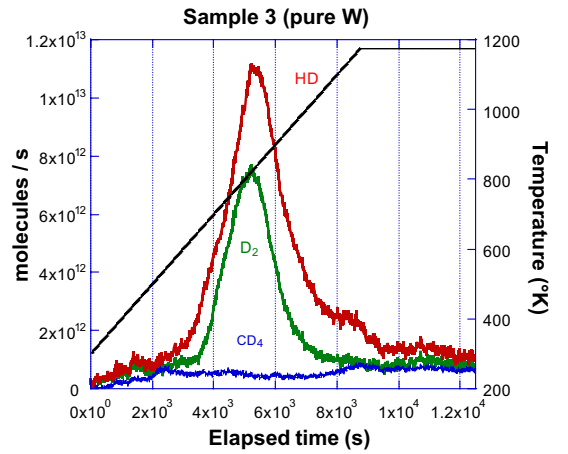

Fig. 4. Flow rate (molecules/s) of D_2, HD and CD_4 as a function of the elapsed time, together with the sample temperature, for sample 3.

between the two samples, but small difference in implantation temperature can lead to very different TDS spectra shape. A comparison was made with TDS results on identical $W + 1\%La_2O_3$ macrobrush samples implanted on the top surface (a factor 4 less large than the two side surfaces in FTU samples) in a deuterium plasma generator with 2.5×10^{22}D ions and plasma electron temperature of 10 eV, at temperature ranging from 773 to 923 K [9]. In that case, no elemental analysis was carried out: annealing before exposure was the same. The TDS spectrum of the sample 4 closely resembles the one of samples implanted at 773 °K (Fig. 3(b)), while the TDS spectrum of the sample 3 is similar to the one of sample implanted at 923 K (not shown for saving space). The results of thermal desorption measurements in terms of total amount of released deuterium (in D atoms), specific amount of desorbed deuterium (in D/m^2) and desorbed/impinging ratio of deuterium atoms are shown in Table 1. Within a factor of three, the desorbed/impinging ratio of deuterium atoms is the same for the two samples, irrespective of the different shape of TDS spectra. The value of this ratio is rather low, not larger than 10^{-4}, as it could be expected on the basis of the very high (close to 1) re-emission coefficient during implantation [7]. On the other hand, if the similarity between spectra shape of Ref. [9] and of present work corresponded at a similarity in average temperature of implantation, we should expect also for FTU samples a maximum specific density of trapped deuterium of about 10^{18}–10^{19} D/m^2, according Ref. [7]. In Ref. [10], for much higher fluence (about 1.0×10^{26} D/m^2) $W + 1\%La_2O_3$ samples exposed at temperature higher than 850 K are reported to reach a retention value of about 2×10^{19} D/m^2. Similarly to samples implanted in the deuterium plasma generator, D retention in macrobrush samples exposed in FTU seems larger than expected: this is even more true if the unknown amount of retained D desorbed during disruptions is taken into account. The larger than expected amount of retained deute-

Table 1
Total and specific amount of released deuterium and desorbed/impinging deuterium ratio for samples 3 and 4

Sample no.	Amount of desorbed D atoms (D)	Specific amount of desorbed D atoms (D/m^2)	Desorbed/impinging ratio (D/D)
3	8.2×10^{16}	7.4×10^{19}	2.3×10^{-4}
4	3.8×10^{17}	3.5×10^{20}	6.2×10^{-4}

rium could be the result of carbon content being large even after plasma exposure. Deuterium could be trapped by carbon itself: in effect, larger deuterium content of C-coated tungsten, with respect to pure W, is to be expected. Nevertheless, it is to be remembered that on FTU samples no carbon coating took place during exposure, but that carbon was already present as carbide. Therefore, a comparison should be done with experiments carried out on carbides: in Ref. [11], deuterium retention in CVD-W_2C is reported to be larger than in CVD-W by a factor of 2. This could not be sufficient to justify the larger than expected D content in our samples. In Ref. [12], carbide formation (WC in this case) is even considered to lead to a reduction in the deuterium retained.

Of course the uncertainty related to the implantation temperature and the real ion fluxes does not allow drawing a definite conclusion. Besides the control of sample temperature and ion fluxes (i.e. by Langmuir probes) during discharges, the comparison between castellated and not castellated clean samples, exposed to nearly identical discharges, could, of course, clarify the matter.

4. Conclusions

Four W and $W + 1\%La_2O_3$ macrobrush samples ($15 \times 15 \times 37\ mm^3$) were exposed to the FTU scrape-off layer plasma, with leading edge flush with the LCFS or up 1 cm apart.

Within the range of EDX analysis (about 0.5 μ), 'as received' samples contained large quantities of O and C, probably as a consequence of machining. The samples were exposed to both steady state and (unintentional) disruptive discharges.

Macroscopic damages include melting and cracks, but no arcing tracks were observed. Gaps are not closed by melted material.

SEM analysis showed patterns of submicron granules on surfaces close to melted zones: due to their bad thermal contact with the material bulk, they can represent an impurity source. Post-exposure EDX analysis showed that O disappeared everywhere huge sample heating occurred.

Thermal desorption spectroscopy carried out on sample 3 (pure W) and 4 ($W + 1\%La_2O_3$) measured total deuterium content of 8.2×10^{16} and 3.8×10^{17} atoms, respectively, corresponding to specific density of retained deuterium of 7.4×10^{19} and 3.5×10^{20} atoms m^{-2}, respectively. The uncertainty in the evaluation of the ratio between retained and impinging deuterium mainly depends on calculated ion flux to the sample.

The deuterium content of the samples seems to be larger than the expected one, as previous tests on macrobrush samples implanted in a deuterium plasma generator suggested. For confirming this impression, comparative experiments on castellated and not castellated samples or reliable localised measurements on exposed and shadowed faces of the same sample are needed.

Acknowledgement

This work was carried out under EFDA Task TW3-TPP-ERTUBE.

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