

Surface modification and hydrogen isotope retention in CFC during plasma irradiation in the Tore Supra tokamak

L. Begrambekov ^{a,*}, C. Brosset ^b, J. Bucalossi ^b, E. Delchambre ^c, J.P. Gunn ^b,
C. Grisolia ^b, M. Lipa ^b, T. Loarer ^b, R. Mitteau ^b, P. Moner-Garbet ^b,
J.-Y. Pascal ^b, P. Shigin ^a, N. Titov ^a, E. Tsitrone ^b, S. Vergazov ^a, A. Zakharov ^a

^a Plasma Physics Department, Moscow Engineering and Physics Institute, 115409 Moscow, Russia

^b DRFC, Association EURATOM-CEA/DSM/DRFC, Centre de Cadarache, 13108 Saint Paul Lez Durance, France

^c EURATOM/UKAEA Association, Culham Science Center, Abingdon, Oxfordshire OX14 3DB, UK

Abstract

The uniform layer with thickness at least 50–100 μm was found on the CFC tiles from the inboard midplane after more than four years of tokamak operation. The upper part of the uniform layer was amorphous, but at the depth of $\sim 5 \mu\text{m}$ a structure consisting of micro-size regions with aromatic chains located parallel to the surface was found. Gradual transition from uniform layer to underlying CFC structure was observed. The reciprocating material probe was used for installation of CFC samples in the Tore Supra deuterium plasma. The thermal desorption spectra of these samples are compared with the spectra of the samples irradiated in the laboratory stand and with the spectra of hydrogenated carbon film. The peculiarities of hydrogen isotope trapping under plasma irradiation and at the atmosphere are presented and discussed.

© 2007 Elsevier B.V. All rights reserved.

PACS: 81.05.T; 61.16.C; 61.80.-x; 79.20.Rf; 34.50.Dy; 52.55.F

Keywords: Retention; Tore supra; Thermal desorption; Carbon-based materials

1. Introduction

Carbon fiber composites (CFC) are being used in contemporary tokamaks and have been selected as a divertor material for ITER. However published studies of plasma influence on the CFC structure and parameters of hydrogen isotope retention in the tokamak conditions are rare.

Formation of the uniform layer on the surface of one-dimensional CFC WMN-type (PAN fibers) irradiated perpendicular to the fiber axes by helium ions ($E = 800 \text{ eV}$, $j \approx 1 \times 10^{20} \text{ ion/m}^2 \text{ s}$, $T = 800$ and 1250 K) was observed [1]. In [2] semi-spherical formations with a stratified structure were found on the surface of three-dimensional CFC of UAM type (PAN fibers) under irradiation in the plasma accelerator VIKA-93 (energy of plasma beam 30 MJ/m^2 , pulse duration $300 \mu\text{s}$, $T > 2300 \text{ K}$). In both cases surface modifications were considered

* Corresponding author. Fax: +7 495 324 70 24.

E-mail address: lbb@plasma.mephi.ru (L. Begrambekov).

as a result of plasma enhanced recrystallization process.

The hydrogen retention were mainly studied for dense graphites in the ion beam facilities with impinging ion energy not exceeding 1000 eV [3–5]. The data received under such conditions hardly can account for CFC retention capacity in the tokamaks. The retention related studies fulfilled in tokamaks are not numerous [6].

The paper presents the first results of investigations of these subjects in the Tore Supra tokamak (T.S.). The samples of CFC N11 (PAN fibers) were subjected to plasma irradiation with the help of reciprocating sample probe mounted at the fast-scanning probe drive [7]. The probe houses five ‘A’-samples at the electron or downstream side and five ‘B’-samples at the ion or upstream side. Hydrogen retention was studied using thermal desorptional spectrometry.

To investigate influence of the T.S. plasma on the structure of CFC, the tiles from the inboard mid-plane were subjected to SEM and TEM analyses. During more than four years of tokamak operation the total period of helium and hydrogen irradiation of the tile surfaces reached several tens of hours.

2. Hydrogen isotope retention in CFC under Tore Supra plasma irradiation

The main features of retention in samples ‘A’ and ‘B’ appear to be quite similar. Thus the paper concentrates mainly on samples ‘B’ (Table 1).

After irradiation in T.S. the samples were kept at the atmosphere for about half a year and then were studied by TDS.

The H₂, HD, D₂ molecules constituted the major portion of the entire thermal desorption. Hydrogen desorption from each ‘A’ and ‘B’ samples was almost the same with the exception of samples B1–B3. Desorption from the latter was half as much. Irradiated surfaces of samples B1–B3 were

Table 1
Parameters of ‘B’ samples irradiation

No.	Fluence (D ⁺ /cm ²)	Energy (eV)	Temperature mean/max (K)
B1	2.3×10^{19}	430	480/750
B2	8.3×10^{18}	290	
B3	2.2×10^{18}	190	450/550
B4	2.8×10^{17}	130	
B5	1.0×10^{16}	60	

found to be covered by semi-transparent ‘rainbow’ layers. These layers disappeared when the samples were heated during TDS investigation.

Deuterium release was much less than hydrogen one for all samples. What is more, HD and in particular D₂ desorption sharply decreased from sample #1 to sample #5. Practically equal amounts of deuterium desorbed in the form of HD and D₂ from samples #1 and #2, but from sample #5 the former was 10 times higher than the latter.

Position of the main peak of H₂-TDS spectra (700–800 K) coincided with that of dense graphite and CFC subjected to low energy (50–200 eV) ion implantation. At the same time remarkable hydrogen release was observed in the low temperature range around 400–600 K and at the temperatures above 1000 K (Fig. 1). Unfortunately, it was quite impossible to carry out correct measurement of TDS spectra at about 1500 K.

The spectra of D₂ and DH desorption from the same samples were rather similar and depended on impinging ion energy (Fig. 2). One should notice a sharp high temperature peak in D₂ and HD spectra.

Analysis of T.S. samples allows conclusively determine only very few features of hydrogen isotope trapping in T.S. samples. In particular it shows: (1) mechanism of hydrogen retention in T.S. samples differs from deuterium retention and (2) there are some relations between ‘rainbow’ layers and particle trapping.

Further investigation was performed in the laboratory stand allowing sample irradiation by ions of gas discharge plasma and TDS-measurement of retained particles [8].

Fig. 3 presents the TDS spectra of H₂ for four samples. Sample #1 was blown around with Ar-

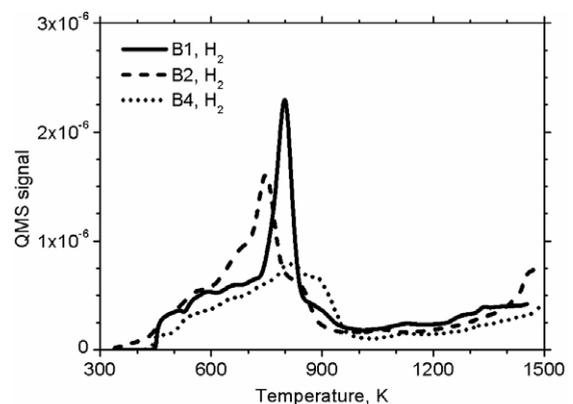


Fig. 1. TDS-spectra of H₂ from samples B1, B2 and B4.

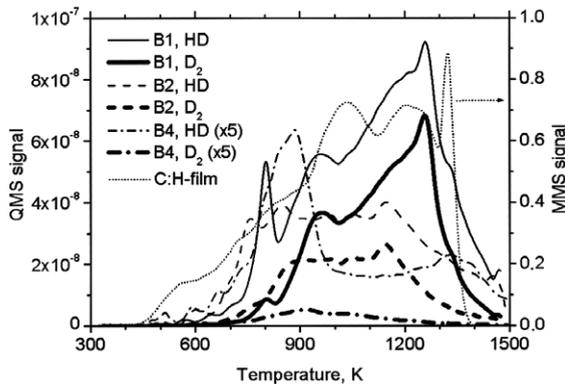


Fig. 2. TDS-spectra of D₂ and HD from samples B1, B2 and B4 and from hydrogenated carbon film.

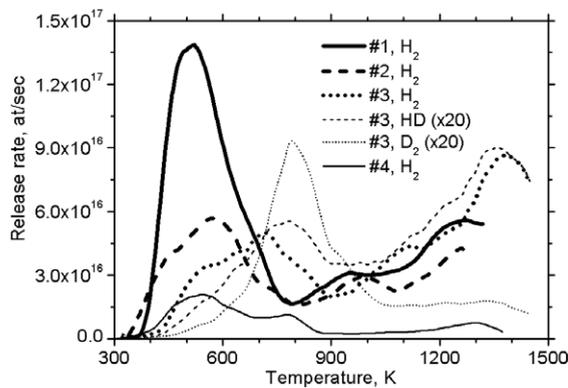


Fig. 3. TDS-spectra of H₂ from samples #1 to #4 and TDS-spectra of D₂ and HD from sample #3.

gas flow. Sample #2 after machining was washed in alcohol in ultra sonic bass in order to remove graphite dust. Sample #3 was treated similarly to sample #1 and then irradiated by deuterium ions ($E_i = 100$ eV/at, $D = 5.4 \times 10^{23}$ at/m², $T = 420$ K). After annealing and keeping at the atmosphere during 30 days first sample was examined as a sample #4.

One can see that the main peak of H₂ spectrum of sample #1 is located in the range of 400–600 K. At the same time remarkable amount of hydrogen is collected in the traps responsible for appearance of the peaks around 900–1000 K and 1200–1400 K. Totally 9.3×10^{22} at/m² of hydrogen are desorbed from the sample including 5.0×10^{22} at/m² in the form of H₂ molecules. The measurement was finished at 1320 K, but some hydrogen might have been collected in traps with higher binding energy.

Hydrogen desorption from sample #2 is much less than from sample #1 which make us think, that

amount of hydrogen collected in graphite dust is practically equal to that in the bulk.

H₂ spectrum of sample #3 demonstrates even stronger decrease of main peak, but high temperature tail of the spectrum stays practically unchanged. It shows that hydrogen collected in the high temperature traps is not affected by ion bombardment.

Sample #4 contains approximately ten times less hydrogen than sample #1. At the same time their TDS spectra are rather similar. The only exception is relatively high peak near 800 K.

Basing on these results one can conclude, that in T.S. samples the main portion of hydrogen was collected in the traps formed during heating and plasma irradiation and devastating around 700–800 K. High temperature traps of T.S. samples were filled by hydrogen before plasma irradiation. The amount of hydrogen in these traps did not depend remarkably on irradiation conditions.

The amount of deuterium desorbed from sample #3 in the form of D₂ (2.8×10^{17} at/m²) and the shape of TDS spectrum with the main peak at 800 K are rather similar to those of preliminary annealed sample. But nearly equal amount of deuterium was additionally desorbed in the form of HD (Fig. 3). The main portion of ‘extra’ deuterium was trapped in the high temperature traps. It might have happened because of isotope exchange process as in case with T.S. samples. One can assume that deuterium retention in high temperature traps iteratively increases along with the increase of irradiation fluence.

High temperature peak of the D₂- and HD-spectra of T.S. sample B1 look similar to the peak appeared in TDS spectra of the hydrogenated carbon film (H:C = 0.5) at the temperature of their destruction (Fig. 2). Thus one can believe, that deuterogenated carbon ‘rainbow’ layers were formed on plasma irradiated T.S. samples B1–B3.

3. Surface modification of CFC tiles during long term T.S. plasma irradiation

The mean plasma flux density on the midplane tiles was estimated to be $(1-2) \times 10^{19}$ at/s m² and the plasma ion energy was in the 100 eV range. The temperature of the tiles reached up to 500–600 °C. More than 20 wall conditioning helium glow discharges were made during this time, resulting in He⁺ ion ($E_i \sim 200$ eV, $j \sim 6 \times 10^{17}$ at/s m²) bombardment of the inner wall.

Continuous uniform layers with stratified structure are found at the surfaces of the examined tiles (Fig. 4). The thickness of the layers is at least 50–100 μm . Cracking and flaking of the upper struts are clearly seen. Flakes of (0.1–0.01) mm^2 are found in the vicinity of overheated leading edges of the tiles. Some fragments of the upper struts have been removed apparently because of stress-induced cracking, holes appearing at the surface. The thickness and volume of the removed fragments reach at least 50 μm and 10^{-3}mm^3 , respectively. The underlying structure of the uniform layer is visible in the remaining holes. The orifices of channels directed into the bulk are seen, apparently at the locations of missing CFC fibers.

Different levels of the transient zone between uniform layer and underlying CFC were observed at small strike area ($\sim\text{cm}^2$) that had been highly damaged by runaway electron impact. In the lowest part of the transient zone many of CFC fibers directed into the bulk disappeared (Fig. 5(a)). The empty rooms are occupied by newly formed column-like

features. They seem to grow from the bulk and have prismatic shapes indicating their crystalline nature (Fig. 5(b)). Linear relief of the prism sides obviously reflects their stratified structure similar to the homogeneous layer. Some parts of matrix have the stratified structure common to both the uniform layer and the new-born columns. In such a way a stratified structure similar to that of the uniform layer penetrates deeply into the bulk CFC. At the periphery of the strike area the upper parts of the transient zone are exposed at the surface (Fig. 6). Concentration of the columns and the modified fragments of matrix are higher over here. The columns and fragments of matrix have rounded tops. The tops merge together and join the nearest edge of the uniform layer.

The upper part of the uniform layer is amorphous with embedded metallic grains. At the depth of $\sim 5 \mu\text{m}$ the layer consists of micro-size regions with aromatic chains located parallel to the surface. They seem to be the first step to formation of pyrolytic structure with basal planes located parallel to

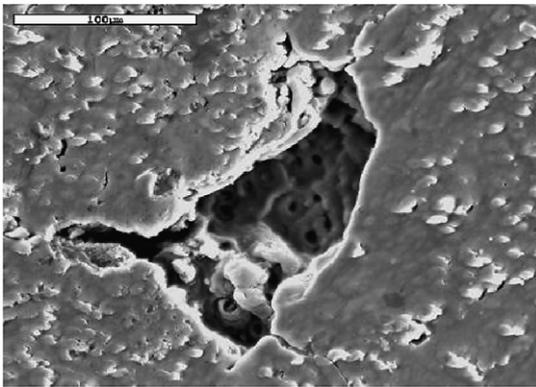


Fig. 4. Continuous uniform layers with stratified structure.

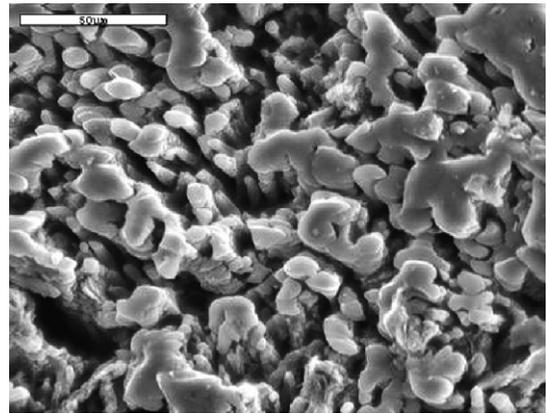


Fig. 6. The upper part of the transient zone.

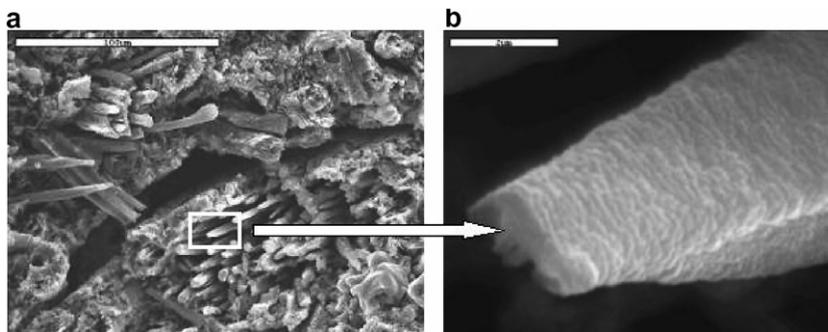


Fig. 5. Transient zone between uniform layer and underlying CFC (a) and newly formed prismatic column (b).

the surface, i.e., destructed surface tends to be crystallized in the structure with minimal free energy. Apparently, formation of the structures with lower free energy could be considered as a general tendency for reconstruction of the surfaces of solids damaged by ion bombardment. It is interesting to note that the same aromatic chain structure was found in the carbon films deposited on the neutralizing plate of the T.S. pumping limiter. Thus, one can believe, that destructed CFC surface and deposited carbon films tend to transform in to the same pyrolytic structure. The redeposited carbon atoms can take part in formation of uniform layer as well.

Formation of homogeneous pyrolytic layer affects parameters of CFC tiles. In particular, the mean surface erosion rate increases as a result of flaking, cracking, emission of the macroscopic particles. Low thermal conductivity of modified layer leads to elevation of the tile surface temperature under high energy particle load. Thus sublimation rate of the surface during disruptions, ELMs, etc., increases and temperature threshold for brittle destruction decreases.

4. Summary

The essential part of hydrogen absorbed in CFC N11 is confined in the high temperature traps. Retention of deuterium into such traps during consequent ion irradiation was much higher than that of annealed CFC apparently due to isotope exchange process. Particle release from these traps starts at the temperature more than 800 K. Assumption is made that retention in CFC can iteratively grow along with fluence due to filling of such traps.

The ‘rainbow’ layers which obviously had deuterogenated structure was found on the CFC surfaces intensively irradiated by T.S. plasma. The ‘rainbow’ layers trap additional amount of deuterium and retain it until the layers are destructed at 1000–1200 K. Hydrogen trapping at the atmosphere also increases at the surfaces covered with ‘rainbow’ layer.

The stratified uniform layer with thickness at least 50–100 μm was found on the CFC tiles from the inboard midplane subjected to more than 4 years of tokamak operation. Wide transient zone was observed between the uniform layer and underlying CFC. The upper part of the uniform layer was amorphous, but at the depth of $\sim 5 \mu\text{m}$ it consisted of micro-size regions with aromatic chains located parallel to the surface. The same structure was found in the carbon films deposited on the neutralizing plate of the pumping limiter.

Homogeneous pyrolytic layers negative influences parameters of CFC tiles.

References

- [1] L.B. Begrambekov, Achievements of science and technology. Charged Beam Particles and Solids, vol.7, Moscow, 1993, p. 4.
- [2] T. Burtseva, I. Hassanein, et al., J. Nucl. Mater. 290–293 (2001) 1059.
- [3] J.W. Davis, A.A. Haasz, D.S. Walsh, J. Nucl. Mater. 176&177 (1990) 992.
- [4] A.Kh. Alimov et al., J. Nucl. Mater. 187 (1992) 191.
- [5] C. Garsia-Rosales, J. Roth, R. Berish, J. Nucl. Mater. 212–215 (1992) 1211.
- [6] T. Takeishi et al., J. Nucl. Mater. 349 (2006) 327.
- [7] J.P. Gunn et al., J. Nucl. Mater. 337–339 (2005) 644.
- [8] L. Begrambekov et al., Phys. Scripta N 108 (2004) 72.