



Analysis and oxidation of thick deposits on TEXTOR plasma facing components

M. Rubel ^{a,*}, J. von Seggern ^b, P. Karduck ^c, V. Philipps ^b, A. Vevecka-Priftaj ^{d,1}

^a *Alfvén Laboratory, Department of Fusion Plasma Physics, Royal Institute of Technology, Association EURATOM - NFR, Teknikringen 31, S-100 44 Stockholm, Sweden*

^b *Institute of Plasma Physics, Forschungszentrum Jülich, Association EURATOM, D-52425 Jülich, Germany*

^c *Central Facility for Electron Microscopy, University of Aachen, D-52056 Aachen, Germany*

^d *Physics Department, Royal Institute of Technology, Stockholm, Sweden*

Abstract

Deuterium and hydrogen containing co-deposited layers formed on plasma facing components at the TEXTOR tokamak were characterised by a number of techniques including surface analysis methods and thermal desorption spectrometry. The aim of the investigation was to determine the composition and structure of the layers prior and after their exposure to air at elevated temperatures of 300°C and 550°C. The impact of the oxidation on the deuterium content and distribution in the surface region and in the bulk of PFC was addressed. The initial deuterium-to-carbon concentration ratio was in the range 0.04–0.06 in the flaking layers on top of the PFC and up to 0.17 on their side surfaces. The oxidation at 300°C for 2 h resulted in partial removal of the deuterium, especially from thin or loosely bound layers, but the release was not accompanied by the change in the deposit structure. The content of plasma impurity atoms in the layers was also not influenced by the exposure to air. Following the oxidation at 550°C partial powderisation of thick, flaking films was observed. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Co-deposition; Tritium inventory; Carbon; TEXTOR; Plasma facing material

1. Introduction

Co-deposition/co-implantation process is a well-known phenomenon occurring on plasma facing components (PFC) in controlled fusion devices [1–10]. The process is a net effect of erosion and deposition: species removed from the wall in one location are transported in the torus and deposited in another, not necessarily distant, location. As a result, hydrogen isotopes are deposited together with plasma impurity atoms forming layers of various structure, composition and thickness

reaching even hundreds of microns. Formation of such films accumulating vast quantities of fuel atoms is considered as a decisive process for tritium retention and inventory in D–T fusion reactors [11,12]. Disintegration of co-deposits into small and loosely bound agglomerates leads to the formation of dust particles in the machines [13]. Safety and economical aspects require the development of techniques resulting in efficient tritium removal accompanied by the decomposition of co-deposited films. Therefore, their properties, i.e. structure, chemical composition and chemical reactivity must be characterised in detail.

The main issues in the study performed were related to: (i) the detailed characterisation of co-deposited films with particular emphasis on the content and distribution of hydrogen isotopes; (ii) the impact of oxidation on the composition and structure of co-deposits, mainly the influence of oxidation on the deuterium and oxygen content.

* Corresponding author. Tel.: +46 8 16 10 61; fax: +46 8 15 86 74; e-mail: rubel@msi.se.

¹ On leave from the Polytechnic University of Tirana, Albania.

2. Experimental

2.1. Deposits studied

The study was carried out with co-deposited layers formed in the TEXTOR tokamak. Two basic types of layers were investigated in detail prior and after their oxidation in air: (i) thick flaking layers formed on plasma facing components such as graphite main poloidal limiter and rf antenna protection tile; (ii) thin co-deposits collected on graphite plates exposed to several tens of discharges.

Figs. 1 and 2 show the appearance of the poloidal limiter and rf antenna protection tile, respectively, after the long term operation of TEXTOR. The base temperature of the tiles was in the range 270–300°C between the shots and exceeding 1000°C during the shots. Though the rf antenna protection tile was located at a radius of 50 cm (in comparison to the minor radius, $a=46$ cm, determined by the ALT II toroidal belt limiter) sharp temperature excursions were observed during some shots due to the plasma positioning. This tile was mounted in the upper part of the protection screen. Both components, the limiter and the protection tile, were installed in the machine several years ago but on the occasion of periodic openings of the vessel the pieces were taken out and their surfaces were cleaned by sandblasting. For the studies reported below, the pieces were removed from the machine in 1997 after the operation period including over 5000 shots fuelled with deuterium, many of them auxiliary heated with NBI and ICRF. The flaking surface layers and the bulk of the tiles were investigated. Examination of the bulk material was preceded by fracturing of the tile and serious precautions were taken to avoid the contamination of freshly open surfaces by deuterium from the outermost layers.

The second category of the studied deposits were thin films formed on graphite plates exposed to the scrape-off

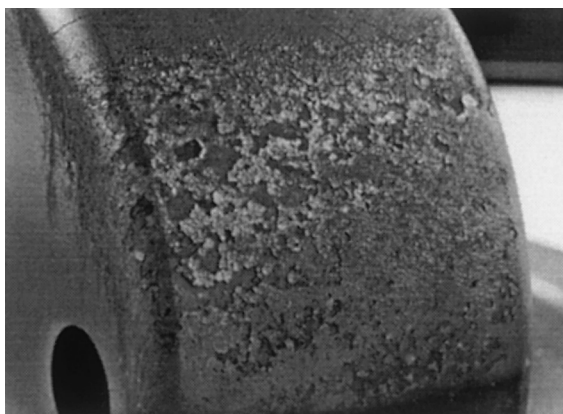


Fig. 1. View of the main poloidal limiter.

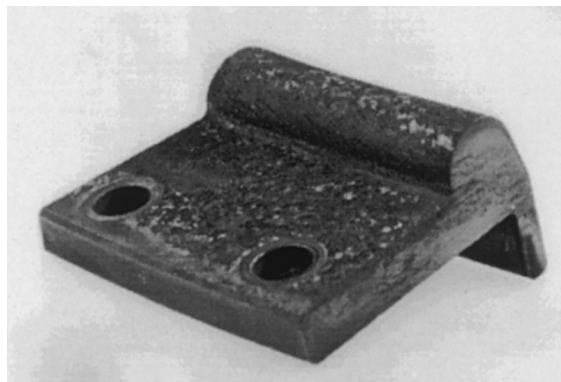


Fig. 2. View of the rf antenna protection tile.

layer plasma. For the exposure, the plates (60 mm long and 18 mm wide) were mounted on the housing of the collector probe. They were exposed to either 24 discharges, including two disrupted ones at the beginning of the experiment, for totally 74.4 s, line averaged density 2.5×10^{19} – $3.7 \times 10^{19} \text{ m}^{-3}$ or to 16 discharges (including two disrupted) for totally 54.2 s, $n_e = 2.75 \times 10^{19} \text{ m}^{-3}$. The plates were at the temperature of 240–260°C between the shots and on some occasions their temperature was exceeding 450°C during the shots ended with disruptions.

The oxidation was performed using a tube oven where selected samples were heated in air at 300–320°C for 2 h and then cooled down to 100°C during another 2 h. The oxidation at approximately 550°C was also performed as well as the heating in vacuum at 300–320°C. Before and after the heating, either in air or in vacuum, the samples were examined by means of various analysis techniques.

2.2. Analysis methods

The main aim was to determine the content on the distribution of hydrogen isotopes in the tiles. Their global contents were measured using thermal desorption spectrometry (TDS). The samples weighing 0.6–1.2 g were heated at $1 \times 10^{-5} \text{ Pa}$ to 1000°C with the heating rate of 26 K min^{-1} . The evolution of masses 1–4 was traced using a calibrated spectrometer, Balzers QMS 420. The depth distribution was inferred from nuclear reaction analysis (NRA) spectra recorded using a $^3\text{He}^+$ beam at 770 (analysis of thin films), 1500 or 1800 keV (analysis of thick films) and detecting protons resulting from the $^3\text{He}(d,p)^4\text{He}$ process. Analyses of plasma impurity atoms co-deposited on PFC was performed with energy dispersive X-ray spectroscopy (EDS), Rutherford backscattering spectroscopy (RBS) and chemical analysis techniques: inductively coupled plasma mass spectrometry and glow discharge mass spectrometry. The

content of boron on surfaces was determined with the NRA technique [$^{11}\text{B}(p,\alpha)^8\text{Be}$] detecting α particles. The topography of the flaking layer, scraped flakes and dust particles was observed with a scanning electron microscope (SEM). In addition, an atomic force microscope was used but the investigation did not provide reasonable data because of the layer roughness.

3. Results and discussion

3.1. Structure and composition of flaking layers

As already shown in Figs. 1 and 2, vast areas of the PFC surface are covered with a non-uniform layer of loosely bound flakes which tend to peel-off. The size of single flakes is from 0.5 to 10 mm, but they are brittle and very easily disintegrate forming much smaller agglomerates of dust particles. Flaking layer can easily be scraped-off even with a plastic tool. However, as can also be perceived, the surface in areas where the flakes peeled-off themselves or were scraped-off is not smooth either showing a tendency to be transformed into the next layer of loosely bound flakes. Some flakes were mechanically scraped-off and their thickness, measured with callipers, was found to be in the range between 80 and 170 μm . The same thickness range was determined from the SEM images of the substrate – deposit boundary recorded for a fractured tile. A distinct gap, up to 15 μm wide, is observed at the boundary. The deposit at the proximity of this gap has a pronounced stratified structure, similar to the one studied by Goodall [9], but the outermost part consists of spherical or oval granules. The top view of the flaking layer is shown in Fig. 3 and the most important feature is a fine structure of the deposit consisting of small (1–20 μm) particles which,

when get detached from the PFC surface, may significantly contribute to the dust formation in a tokamak. There is a wide variety of structures of particle agglomerates which form flakes. The removal of the outermost layer reveals a rough surface with cracks, as exemplified in Fig. 4.

Major constituents of the co-deposited layer are carbon and deuterium, but significant amounts of other species are found because the elemental composition of flaking layers reflects the history of their formation. The characteristic feature for the co-deposits from TEXTOR is the presence of silicon and boron in quantities of a few weight percent. This is due to the wall conditioning by siliconization or frequent boronization, injection of $\text{B}(\text{CH}_3)_3$ or SiH_4 and testing of Si or B doped composite limiters. Large quantities of oxygen detected in the surface region are partly related to the gettering by Si and B in the machine, but the majority of it originates probably from the uptake of oxygen from air during the storage of samples in the laboratory; this issue has been discussed in detail elsewhere [14]. Other plasma impurity atoms detected in the deposits are: Ni, Cr, Fe and Mo (originating from the inconel liner) and Ti, W, S, Cl and Ca. The same species in similar quantities are detected on the tiles' surface exposed after scraping-off the flakes. Microbeam EDS study, probing the near surface layer of less than 1 μm , reveals highly non-uniform areal distribution of elements. For instance, there are spots consisting of either metal, carbon or silicon particles, only.

3.2. Deuterium content and distribution

Fig. 5 and Table 1 present the results for the deuterium content measured with NRA and TDS, respectively, in different parts of the rf antenna protection tile

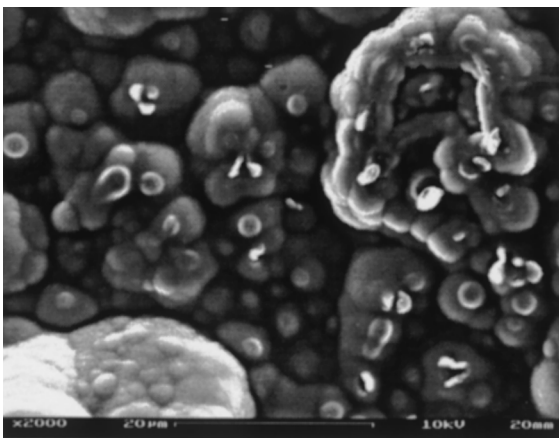


Fig. 3. Surface topography of the flaking layer on the rf antenna protection tile.

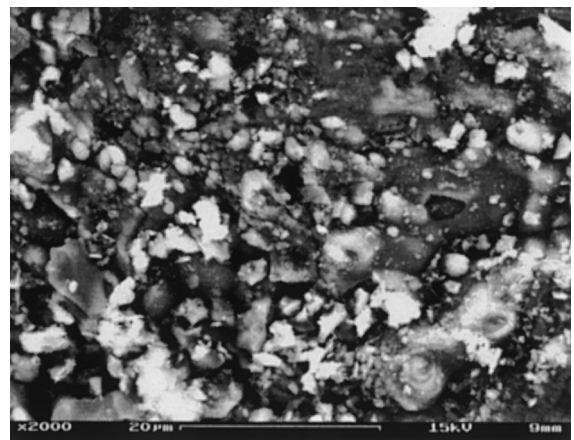


Fig. 4. Surface topography of the graphite substrate after the removal of the flaking layer.

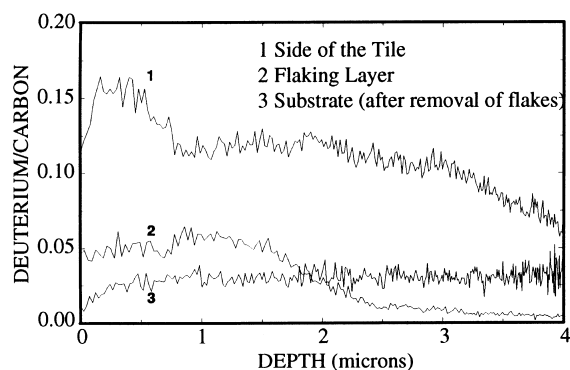


Fig. 5. Deuterium depth profiles determined in different areas of the rf antenna protection tile.

before the oxidation in air. The deuterium-to-carbon ratio as a function of depth is shown in Fig. 5 for three areas: the side of the tile covered with a deposit of smooth appearance (plot 1), the area with thick flakes, (plot 2) and the area from which the flakes were removed by scraping them off (plot 3). The differences between the depth profiles are fairly distinct when the contents and distributions are considered. The greatest D contents, in the range 0.6×10^{18} – 4.7×10^{18} cm^{-2} (4.4×10^{18} cm^{-2} in the case illustrated by plot 1), are observed on the tile's side, i.e. in the area hidden in a gap between the antenna protection pieces. This surface was not as hot as the outer surfaces were. This is in a qualitative accordance with previous results, for instance from JET, showing the greatest deposition on the side surfaces of the divertor tiles [4]. The layer shown in plot 1 has a maximum depth of about 5 μm as inferred from the NRA-based depth profiles recorded with the increasing energy (1500–1800 keV) of the analysing He-3 beam. This statement is also supported by the fact that the D contents measured both by means of TDS and NRA, in the selected area of the side surface, are in a good agreement, as shown in the second row of Table 1.

On the surface covered with thick flakes the areal distribution of D atoms is fairly uniform, 0.9×10^{18} – 1.2×10^{18} cm^{-2} , in the depth of 4–5 μm accessible with NRA. This has been proven by measurements per-

formed in more than 20 different points, separated by a few millimetres one from another, in order to obtain decent statistics and to decrease the possibility of misinterpretation of the results obtained for this rough surface. Plot 2, which is representative for the spectra recorded, shows also that the deuterium depth distribution in the flaking layer is highly inhomogeneous with the majority of atoms accumulated in the near surface region 1.5–2 μm thick. From these measurements alone one cannot judge about the distribution and content of deuterium in deeper region of the flakes, but the TDS analysis of the sample with the flaking layer gives the total content of 2×10^{18} cm^{-2} which is only by a factor of 2 greater than the near surface concentration. Following the mechanical removal of flakes the D content determined with NRA on the graphite substrate is about 1×10^{18} cm^{-2} (plot 3). Therefore, the sum of the concentrations measured with NRA in the flaking layer and in the substrate is about 2×10^{18} cm^{-2} . This value is equal to the total D content determined with TDS for the piece covered with flakes. On this basis one may suggest that the flakes on the investigated PFC contain totally about 1×10^{18} cm^{-2} of deuterium accumulated mostly in the surface region, 1.5–2 μm in thickness. It is plausible, therefore, that the deuterium from the deeper region was released during the earlier stages of the plasma operation, i.e. during the earlier stages of the deposit growth. It is also noticeable that the deuterium-to-carbon ratio does not exceed the value of 0.17 on the side surfaces and 0.06 in the flaking layer. Most probable explanation of these fairly low values is related to the fact that the base temperature of the PFC was 270–300°C and much higher temperatures (over 1000°C) were measured during shots and, as a consequence, it stimulated quite an efficient release of deuterium from the tiles already in the machine.

TDS measurements show that the analysed pieces covered with a deposit also contain remarkable quantities of hydrogen which are by a factor of 1.5–3 greater than the deuterium quantities (see rows 1 and 2 in Table 1). This is probably due to the water adsorption when the samples are stored in air before the TDS analysis and, to some extent, due to the H–D isotope exchange and diffusion of hydrogen into the bulk. The statement is partly justified by the fact that the hydrogen is also detected, though in 4–5 times smaller quantities,

Table 1
Deuterium and hydrogen contents in different parts of the rf antenna protection tile

Sample	Sample mass (g)	Deposit area (cm^2)	C_D (TDS) (cm^{-2})	C_H (TDS) (cm^{-2})	C_D / C_H	C_D (NRA) (cm^{-2})
Front area with flakes	1.2880	2.40	2.0×10^{18}	3.0×10^{18}	0.67	1.1×10^{18}
Side of the tile: smooth deposit	1.2264	1.41	0.78×10^{18}	2.3×10^{18}	0.34	0.9×10^{18}
Bulk material	0.6120	–	9.4×10^{15} (cm^{-3})	0.55×10^{18} (cm^{-3})	0.02	–

in the sample of the bulk material. The content of H is also by a factor 50 higher than that of deuterium in the same sample. Moreover, the bulk content of D determined with TDS is below 1 appm and there is an agreement between this result and the one obtained with the NRA method on the surface freshly open by cleaving the tile.

3.3. Oxidation of co-deposits

The samples of PFC were exposed for several hours to air at the temperature of either 300–320°C or 550°C in order to assess the rate of D removal and the decomposition of co-deposits in oxidising atmosphere, i.e. the oxidation of hydrogen isotopes to water vapour (HDO) and the oxidation of carbon to its oxides. The oxidation at 300–320°C resulted in the decrease of the deuterium content by a factor of 3 from 0.91×10^{18} to $0.30 \times 10^{18} \text{ cm}^{-2}$. This decrease was not accompanied by the change either in the shape of the depth profile (in comparison to the one shown in plot 2 of Fig. 5) or in the layer structure which remained basically the same as shown in Fig. 3. No significant deuterium release was noticed from the surface from which the flakes were mechanically removed prior to the oxidation; the D content and the surface structure remained unchanged and only a small change in the depth profile occurred in comparison to the one illustrated by plot 3 in Fig. 5. These apparently contradicting results may be caused by the differences in the surface structure of the two samples: (i) loosely bound, thin, very porous flakes; and (ii) a thick, solid substrate. One may suggest that the more developed surface area of the flaking layer with deuterium accumulated mainly in the outermost region enables better penetration of oxygen to the film and it results in a higher removal rate of deuterium from flakes than from the solid substrate.

To check the influence of oxygen on thin films, the oxidation of deposits (80–100 nm) collected on graphite

plates was performed. The deposit was collected in TEXTOR a few years ago and the D content was measured afterwards. The second measurement was done five years later (just prior to the oxidation) and the third one after the exposure to air at 300°C. The results for those three cases are in Fig. 6 showing that: (i) during the five years storage approximately 30% of deuterium was released, probably due to the isotope exchange; (ii) the oxidation resulted in the removal of about 80% of the remaining deuterium. However, as in the case of thick co-deposits, no change was noticed in the appearance of the sample. The colour pattern of the interference fringes and the content of co-deposited silicon atoms were the same in all three cases. The observations may indicate that the deposit thickness was not distinctly reduced during the oxidation. There were also no significant differences observed in the oxygen content in the co-deposits before and after their oxidation showing that the layer was saturated with oxygen already before the heating in air. The result confirms the stability of co-deposits' structure under the oxidation at 300°C and it also indicates relatively efficient, however, not rapid oxygen-stimulated removal of deuterium from this layer.

The oxidation in air during 2 h at 550°C resulted in the removal of 95% of the deuterium accumulated in the flaking layers. This process was accompanied by partial powderisation of the film. From the sample with mechanically removed flakes (prior to the oxidation) about 85% of deuterium was released.

4. Conclusions

Summarising the experimental results one may tentatively conclude that the content of hydrogen isotopes (H and D) on the surfaces and in the bulk of PFC kept in TEXTOR at the base temperature 270–300°C does not exceed the level of $5 \times 10^{18} \text{ cm}^{-2}$. However, one cannot decisively conclude on the total amount of fuel atoms being accumulated in co-deposits during the whole operation period when the PFC were in the machine. This is because the flaking layers might easily peel-off and fall down to the reactor floor in the form of dust particles containing also fuel species.

The exposure of thick co-deposits and flaking layers for several hours to air at the temperature of about 300°C results in the partial release of deuterium. This process is not accompanied, however, by the change in the structure of the surface layers, indicating that the deposit itself is not rapidly removed or decomposed and retains its porous structure. Therefore, it probably may efficiently trap fuel atoms again during the plasma operation following the oxygen-assisted clean-up procedure. The study of this issue is under way.

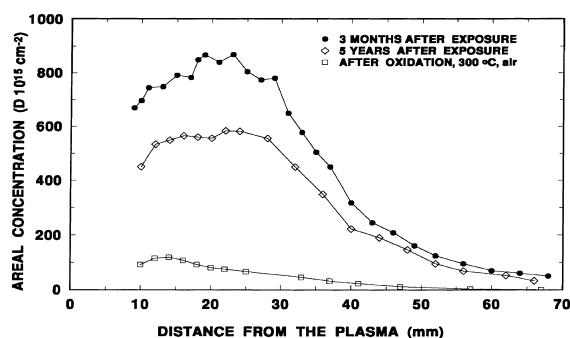


Fig. 6. Deuterium content in thin co-deposits before and after their oxidation in air.

Acknowledgements

The Wallenberg Foundation is highly acknowledged for funding the SEM and EDS equipment. The work was partly performed under the NFR Contracts F-AA/FU 06571-317 and F-FF 06571-321.

References

- [1] J.P. Coad, B. Farmery, *Vacuum* 45 (1994) 439.
- [2] R. Behrisch et al., *J. Nucl. Mater.* 145–147 (1987) 723.
- [3] B.E. Mills, D.A. Buchenauer, A.E. Pontau, M. Ulrickson, *J. Nucl. Mater.* 162–164 (1989) 343.
- [4] J.P. Coad, M. Rubel, C.H. Wu, *J. Nucl. Mater.* 241–243 (1997) 408.
- [5] R. Causey, W.R. Wampler, D. Walsh, *J. Nucl. Mater.* 176&177 (1990) 987.
- [6] M. Rubel, H. Bergsäker, P. Wienhold, *J. Nucl. Mater.* 241–243 (1997) 1026.
- [7] P. Franzen et al., *Nucl. Fusion* 37 (1997) 1375.
- [8] A.P. Martinelli, A.T. Peacock, R. Behrisch, *J. Nucl. Mater.* 196–198 (1992) 729.
- [9] D.H.J. Goodall, *Contr. Workshop on Plasma–Wall Interactions at JET, Abingdon, UK, 1987.*
- [10] N. Almqvist, M. Rubel, S. Fredriksson, B. Emmoth, P. Wienhold, L. Ilyinsky, *J. Nucl. Mater.* 220–222 (1995) 917.
- [11] G. Federici, D. Holland, G. Janeschitz, C.H. Wu, *J. Nucl. Mater.* 241–243 (1997) 260.
- [12] G. Federici et al., *these Proceedings.*
- [13] J. Winter, *these Proceedings.*
- [14] P. Wienhold, M. Rubel, J. von Seggern, H. Künzli, I. Gudowska, H.G. Esser, *J. Nucl. Mater.* 196–198 (1992) 647.