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Ion-induced release of deuterium from co-deposits by high energy helium bombardment

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Abstract

Ion-induced release of deuterium from thick and thin co-deposits formed on plasma facing surfaces in a tokamak and from thick hydrogenated carbon films by MeV $^3He^+$ beams was studied. The layers of different thickness and D content, up to 2×10^{19} cm^{-2}, built on graphite, CFC, Ni, Ti, V and Mo under different operation conditions (i.e. heating mode or wall composition in the machine) were irradiated with ^3He ions in the MeV range, 0.8–1.5 MeV and a total fluence exceeding 1×10^{17} He^+ cm^{-2}. The D content was determined by nuclear reaction analysis (NRA) and the films, prior and after the irradiation, were also examined by RBS and EDS. The amount released was found to be dependent on the layer elemental composition: 55–60% from the carbon and boron containing films and only 10% from those containing silicon. Effective detrapping cross-sections have been calculated on the basis of the results obtained.

Keywords: Wall particle retention; Erosion and particle deposition; TEXTOR

1. Introduction

Co-deposition of hydrogen isotopes together with plasma impurity atoms, especially carbon, on plasma facing components in controlled fusion devices is considered as a decisive process for tritium retention and inventory. It also decides recycling properties of hydrogen and, eventually influences the plasma density control in tokamaks. It has been shown that thick co-deposits, with high H isotopes content, are formed on surfaces located in the deposition zones; for instance, on certain areas of limiters, divertor and rf antenna protection tiles [1–4]. The built-up of co-deposits, exceeding 6–7 μm in thickness and containing above 1×10^{19} D atoms cm^{-2}, has also been observed on sides of the brush-type divertor tiles i.e. on the surfaces not facing the plasma directly [2]. In addition, the concern that hydrogen accumulated in the layers may migrate into the bulk of carbon based materials has lead to extensive studies of the phenomena [5–7]. Very extensive investigation of co-deposits is carried out in order to recognize both their morphology (structure, composition [8–11]) and behaviour under thermal treatment [7,12]. The major issue is to assess the conditions leading to the layer decomposition and hydrogen removal. Comparative studies of thermal release performed for hydrogenated carbon films prepared by deposition in laboratory plasma facilities have shown similar hydrogen desorption characteristics as tokamak co-deposits. However, the layers of both kinds are not identical. They have different microstructure and the overall elemental composition because tokamak co-deposits contain not only C and D but also many plasma impurity atoms, for instance B, Be, Si, medium and high Z metals. The same applies to hydrogenated layers prepared by direct H or D implantation [13–16], because of fairly localized deposition of implants in relatively thin layers (not exceeding hundreds of nm), in contrary to the co-deposits which can be much thicker with broad and not necessarily uniform distribution of hydrogen isotopes.

It is also known that the irradiation of C–H layers with a MeV He^+ beams results in the detrapping of D as studied in detail by Roth et al. [13] who discussed the detrapping mechanism and calculated empirical cross-sections. Similar results have been obtained by Morita and

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Hasebe [17] using the ERD technique. Bergsåker et al. [18] observed differences in the D release characteristics from a thin tokamak co-deposit in comparison to hydrogenated films prepared under laboratory conditions. However, still relatively little is known about detrapping from thick tokamak co-deposits, especially about the change in the depth distribution of D under the MeV He irradiation.

This contribution reports on the ion stimulated detrapping of deuterium from co-deposited layers containing mostly carbon, but also significant quantities of other atoms like boron or silicon. The aim of the investigation was to determine the efficiency of deuterium release. Two issues were of particular interest: (i) the detrapping from thick co-deposits and (ii) the influence of the co-deposit elemental composition, (related to the wall composition in the machine during the deposit formation) on the release characteristics.

2. Experimental

The investigation was carried out with deuterium containing co-deposited layers formed on plasma facing surfaces at the TEXTOR tokamak. For comparison, amorphous hydrogenated carbon films produced in laboratory plasma facilities were studied. For the exposure at TEXTOR, the substrates were attached on the collector probe transfer system. They were facing the scrape-off layer plasma during 4–120 s corresponding to 1–34 pulses (Ohmic or additionally heated with ICRF or NBI) fuelled with deuterium. Series of exposures were made with different collecting substrates and under different wall conditions (carbonized, boronized and siliconized) in order to study the influence of the collecting substrate and the layer composition (C, B and Si) on the retention of hydrogen and ion induced removal. The thickest deposit (of those reported below) was formed on the C and Al substrates facing the plasma for only 34.8 s in the following sequence of discharges: two Ohmic, five with the NBI co-injector (1.6 MW) followed by two pulses heated with neutral beams with the co- and counter injectors. The exposure was made in the boronized TEXTOR operated with a test carbon limiter doped with 3% of boron. The test limiter was in the same radial position, $a = 46$ cm, as the blades of ALT II pump limiter, the main one in operation.

Another set of films was prepared by exposing carbon-based substrates to a deuterium plasma in a hollow cathode; the total fluence of deuterium ions to the target was 1×10^{21} cm^{-2} and the targets' temperature during the exposure was below 70°C.

The deuterium content on the plasma exposed surfaces was then examined with nuclear reaction analysis (NRA) using a 770 keV or 1500 kev $^3He^+$ beam and detecting protons emerging from the $^3He(d, p)^4He$ process. The amount of D retained was controlled during the prolonged irradiation (flux density $0.8–1.5 \times 10^{13}$ cm^{-2} s^{-1} and the total $^3He^+$ fluence up to 1×10^{17} cm^{-2}) accompanied by the deuterium detrapping. Rutherford backscattering spectroscopy (RBS) was applied to determine the quantity of heavy plasma impurity atoms in the co-deposits (Si and metals), nuclear reaction $^{11}B(p, \alpha)^8Be$ for boron and enhanced proton scattering $^{12}C(p, p)^{12}$ for carbon determination on non-carbon substrates.

3. Results and discussion

3.1. Tokamak co-deposits

Fig. 1a–d show the sequence of the NRA spectra for deuterium in the deposits collected on graphite and alu-

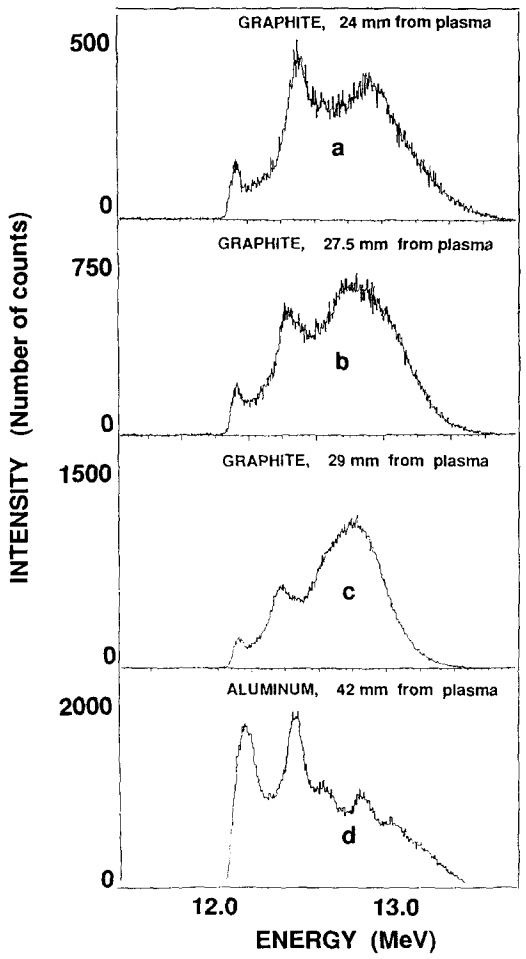

Fig. 1. NRA spectra for deuterium deposited on graphite and aluminum substrates facing the scrape-off layer plasma at TEXTOR: (a) 24 mm; (b) 27.5 mm; (c) 29 mm and (d) 42 mm in the limiter shadow.

minum substrates exposed for 34.8 s to the scrape-off layer plasma: from 24 mm up to 42 mm in the shadow of the limiter. On the spectra, the intensity is plotted versus the energy of detected particles. The total D concentrations measured for the layers were: (a) 1.53×10^{18}, (b) 1.88×10^{18}, (c) 2.57×10^{18} and (d) 2.75×10^{18} cm^2. The most striking features are: (i) the significant difference in the shape of the spectra accompanied by the differences in the deuterium content, suggesting drastic changes of the deuterium distribution through the layer and (ii) the increasing deuterium amount radially from the plasma. Exponential decrease in the deposition profile was observed from 51 mm deep in the SOL.

The depth distribution of the deuterium deposited and its ratio to carbon (C_D/C_C) is exemplified in Fig. 2 for the layer collected 29 mm from the plasma edge. Plots confirm highly stratified structure of the layers and one can distinguish at least three maxima in the spectra. The co-deposit thickness reaches 4 μm and the D distribution is highly non-uniform with the highest content at the depth of approximately 3 μm. One may suppose that the non-uniformity of deuterium distribution in the layer is strongly related to the history of the layer formation during tokamak pulses, as described in the previous section. NBI heated pulses, including the two last with both injectors in use, probably resulted in the formation of fairly thick deposits (carbon and boron) but retaining relatively low amount of deuterium which was strongly released thermally from the deposit already during the shots. This statement is justified by the pronounced temperature increase (above 500°C) of the probe during NBI pulses, whereas no significant temperature rise was observed in Ohmic discharges; a thermocouple was mounted at the end of the probe. As a result, the C_D/C_C ratio in the outer region (depth up to 1.5 pm) of the deposit is much lower than in the deeper layers. Those deeper layers were also affected by thermal loads, because the C_C/C_D value in that region does not exceed 0.11. Therefore, it is nearly four times lower than the value around 0.4 frequently observed for tokamak co-deposits. Moreover, even lower values (0.08 and 0.06, at the maximum), have been found in the deposits collected closer to the plasma (27.5 and 24 mm from its edge, respectively, features are shown in Fig. 1a, b) whereas $C_D/C_C = 0.21$ has been detected at the more distant position (Fig. 1d) on the aluminum substrate as determined in direct measurements of C and D deposited (carbon analysis using the $^{12}C(p, p)^{12}C$ reaction). In the latter case the highest amount of deuterium has been detected at the near surface region. One may suggest that the different structure of the co-deposits in Fig. 1a–d are mostly connected to the target temperature during the exposure (highest probe temperature close to the plasma), but rather not to the substrate composition. It has been shown [19] that there is no significant influence of the substrate composition on the collection efficiency of co-deposits.

The three plots (a–c) in Fig. 2 correspond to three different stages of the layer irradiation with a $^3He^+$ beam: 4.68×10^{14}, 23.4×10^{15} and 46.8×10^{15} cm^{-2}. They are recorded for the co-deposit shown in Fig. 1c. These depth profiles prove a gradual but substantial release of deuterium under the helium bombardment. Prolonged irradiation reduces the concentration from 2.57×10^{18} cm^{-2} to 1.4×10^{18} cm^{-2} corresponding to the release of 46% of deuterium from the layer. However, the concentration drop is not homogeneous through the layer. In the near surface

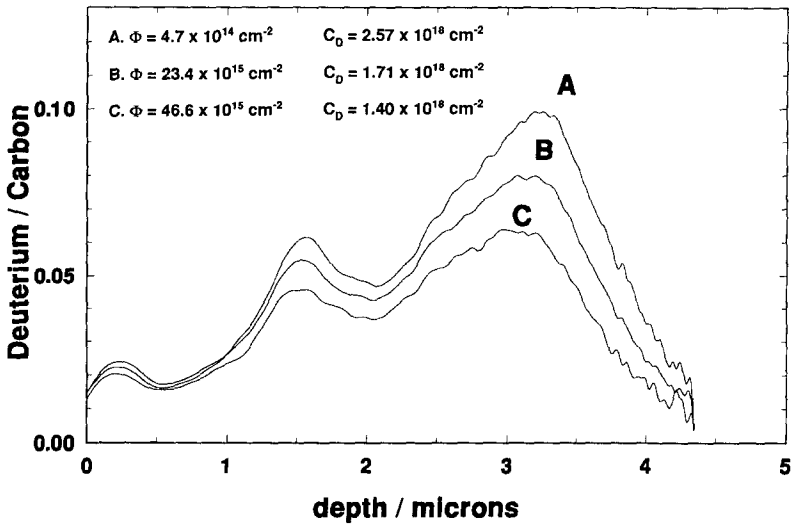

Fig. 2. The change in the deuterium depth profiles following the layer irradiation with an increasing fluence of 1500 keV ^{8}He ions. The profiles are recorded for the co-deposit shown in Fig. 1c.

region the concentration remains nearly unaffected whereas the released amount increases with depth. In more quantitative terms: 51% is released from the most deep region of the deposit; 34% from the middle layer (0.5–1.5 μm) and less than 5% from the surface region. This detrapping characteristic may be attributed to several factors related both to the nature of the detrapping process itself and to the nature of hydrogenated co-deposits containing different C–H binding states. The detrapping of deuterium by MeV He ions occurs mostly via the electronic excitations [18] and the effect is pronounced at a greater depth, where the ion energy is deposited most effectively. The electronic stopping power of 1500 keV ^{3}He ions in carbon increases with depth reaching its maximum between 3 and 4 μm (see the ^{3}He stopping power profile included in Fig. 3). Secondly, hydrogenated carbon films retain deuterium in states of different binding energy as discussed by Causey et al. [20]. The variety of binding states may exist in C–H films with high content of hydrogen isotopes. Loosely bound states are easily decomposed. It may explain the fact that relatively more deuterium is released, and more easily, from the depth of the deposit than from the surface layer containing small amounts of D. The surface layer is probably less abundant in low energy binding states because of the thermal effects (the rise of the probe temperature by a few hundreds degrees during NBI pulses), as mentioned above.

Released atoms, before recombination, molecular diffusion and desorption from the surface [21], may be re-trapped in the film. The efficiency of this process is known to be proportional to the availability of free traps, i.e. inversely proportional to the D content. In the layers under investigation, the highest re-trapping probability occurs in the surface layer. It may explain why the surface concentration remains nearly constant during the irradiation: atoms released are replaced by the detrapped ones and there is no net effect observed for the concentration change.

The results described above have been obtained for the deposits formed at TEXTOR operated with the boronized wall and a boron-doped carbon test limiter. Comparative measurements of the detrapping characteristics were also performed for thin co-deposits formed in the presence of carbonized, boronized and siliconized walls. The deposits containing 0.6–1.0 $\times 10^{17}$ D atoms cm^{-2} (thickness 40–60 nm as determined by colorimetry) were collected during Ohmic discharges. No significant temperature rise of the collecting substrates was observed within the exposure. The efficiency and the rate of the deuterium release from films formed in the presence of the carbonized and boronized walls were nearly the same and up to 55–60% of deuterium was detrapped with ^{3}He$^+$ fluences of 6 $\times$ 10^{16} cm^{-2}. In contrary, only 10–12% of D could be removed from the co-deposits collected very shortly after the siliconization of TEXTOR; the collected co-deposits contained in this case mainly silicon and only traces of boron. The release occurred only during the initial phase of irradiation with the dose not exceeding 5 $\times 10^{15}$ cm^{-2}. This confirms high stability of hydrogen in the Si-containing layers.

The data presented for tokamak co-deposits prove a big variety of the deposit properties dependently on the operation conditions in the machine. They also prove that the C_D/C_C ratio may vary over wide range of values, especially in thick layers.

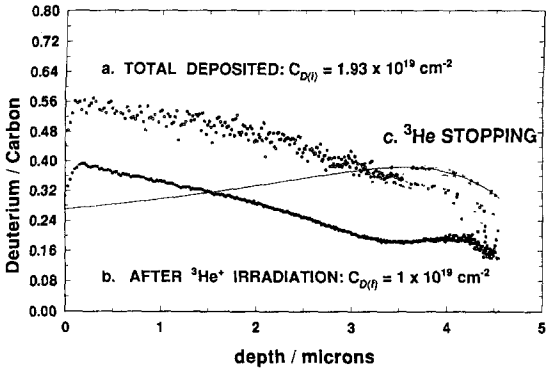

Fig. 3. Deuterium depth distribution and ^{3}He+ induced detrapping from the amorphous hydrogenated film formed in a hollow cathode discharge; (a) helium fluence 2.34$\times 10^{15}$ cm^{-2}; (b) helium fluence 1.45$\times 10^{17}$ cm^{-2}; (c) ^{3}He electronic stopping power in carbon.

3.2. Hydrogenated carbon films

Fig. 3 shows the depth profiles of deuterium in the film produced under mild conditions (temperature below 70°C) in a hollow cathode discharge in the D plasma. The analysis was performed a few days after the exposure and the deuterium content was fairly uniform over the exposed surface. The profiles correspond to (a) the film in the initial stage of irradiation with helium and (b) after bombardment with a dose exceeding 1 $\times 10^{17}$ cm^{-2}. In addition, the electronic stopping of He in carbon is plotted to illustrate its change versus depth. The thickness of the film is at least 4 μm (the limit of the detection depth with a ^{3}He$^+$ beam at 1500 keV). The film is highly saturated with deuterium and the C_C/C_D ratio distinctly exceeds the value of 0.4; total deuterium deposited 1.9 $\times 10^{19}$ cm^{-2}. Following the helium bombardment the deuterium content drops by 48% to the level of 1 $\times 10^{19}$ cm^{-2}. In contrary to the profiles for thick tokamak co-deposits, the depth profile in Fig. 3 is not changed by the irradiation. The concentration decrease is fairly monotonic over the whole layer. It may be due to the very high overall content of D (probably over-saturation) and therefore a limited number of traps for the detrapped atoms.

There are pronounced distinctions, in the structure and also in the C_D/C_C ratio, between thick tokamak co-deposits and layers formed in laboratory plasma facilities. That is caused by different conditions, for instance temperature and presence of tokamak plasma impurities, accompanying the layer growth. Therefore, the results obtained indicate also how difficult (or irrelevant even) may become direct comparisons of layers formed under the tokamak plasma discharges with those growing under mild conditions.

4. Summary

The studies performed show significant differences in the detrapping characteristics of deuterium from co-deposits dependently on the layer composition i.e. on the conditions accompanying the layer formation. From most of the co-deposits up to 55–60% of deuterium can be removed by the He^+ bombardment and the highest removal rate is observed at the initial stage of irradiation, He^+ dose up to $6–10 \times 10^{14}$ cm^{-2}. The lowest efficiency (10% maximum) is found for the Si containing layers and also for the pre-heated (550°C) films.

It has been observed that during the D–T operation there is small flux (5% or less) of unconfined helium [22]. Therefore, one may expect that even low fluences of unconfined α particles heating the tokamak wall will probably stimulate fairly efficiently in-situ tritium removal from the co-deposits formed during the D–T operation. Detrapping from beryllium-containing layers is under investigation.

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