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Laser-induced removal of co-deposits from graphitic plasma-facing components: Characterization of irradiated surfaces and dust particles

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ABSTRACT

Laser-induced fuel desorption and ablation of co-deposited layers on limiter plates from the TEXTOR tokamak have been studied. Gas phase composition was monitored in situ, whereas the ex situ studies have been focused on the examination of irradiated surfaces and broad analysis of dust generated by ablation of co-deposits. The size of the dust grains is in the range of few nanometers to hundreds of micrometers. These are fuel-rich dust particles, as determined by nuclear reaction analysis. The presence of deuterium in dust indicates that not all fuel species are transferred to the gas phase during irradiation. This also suggests that photonic removal of fuel and the ablation of co-deposit from plasma-facing components may lead to the redistribution of fuel-containing dust to surrounding areas.

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1. Introduction

Development of methods for fuel removal and decomposition of co-deposited layers from plasma-facing components (PFC) is an urgent task to control and mitigate tritium inventory in next-step fusion devices. The complexity of this task has been understood leading to the conclusion that no single method alone – proposed until now [1] – would be fully sufficient to solve the problem. Among several techniques being developed for that purpose, photonic cleaning by laser pulses has been tested on PFC in laboratories [2–7] showing that the whole co-deposited layer could be removed [8–10]. The efficacy of fuel removal by flash-lamp light was tested in the JET tokamak during a shut-down period using a remote handling system [11]. To characterize fully the process of cleaning it is necessary to diagnose the products generated by irradiation: gas phase composition and dust. In situ optical spectroscopy has shown the decrease of deuterium in the gas phase during the removal process [7,12,13]. The main subject of this paper is focused on the collection and characterization of dust and laser-treated surfaces.

2. Experimental

The study of fuel and co-deposit removal has been performed in laboratory on a limiter plate retrieved from the TEXTOR tokamak after plasma operation period of 4 h. A tile from the toroidal belt pump limiter (ALT-II, Advanced Limiter Test II) made of graphite (Toyo Tanso, IG-430U) was used as the target for irradiation with laser pulses. ALT tiles have two distinct regions: net erosion and net re-deposition zones [14,15]. The irradiation was done on the re-deposition zone covered by 40–50 μm thick co-deposits. Their properties such as structure, elemental composition and fuel content have been earlier reported in detail [14,16].

Two types of experiments have been performed: laser-induced ablation of co-deposits (IPPLM Warsaw) and laser-induced desorption of fuel (at Forschungszentrum Jülich, FZJ). A schema of the experimental set-up used for laser-induced ablation is shown in Fig. 1(A); more details can be found in [6,7]. Single shots and series of laser pulses (up to 200) were performed using a repetitive a Nd:YAG laser (NL303HT model from EKSPLA) delivering 3 ns pulses of energy of up to 0.8 J and adjustable repetition rate of up to 10 Hz at the wavelength of 1060 nm. The laser beam had the "top hat" spatial beam profile and divergence less than 0.5 mrad. The convex lens of a focal length of 70 cm was mounted outside of the vacuum chamber at the distance of 45 cm. This ensured optimal beam focusing at the target as proven in previous experiments where a wide range of lens-target distances were tested to estimate the

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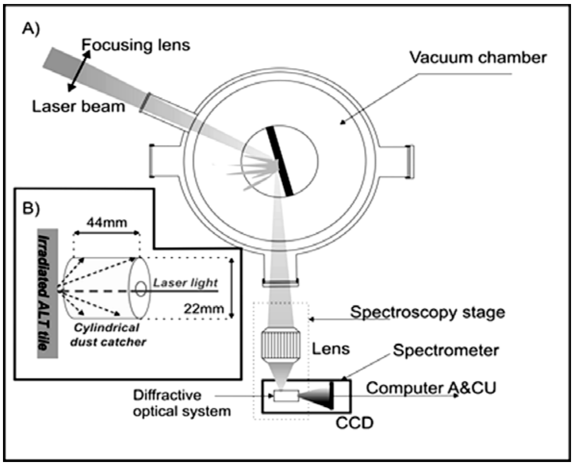

Fig. 1. Scheme of the experimental set-up (A) and cylindrical dust catcher (B).

threshold values for ablation of the co-deposit and bulk graphite [6]. The angle between the beam and normal to the target was about 20, so the beam spot on the target had ellipsoidal shape with semi-major and semi-minor axes of around 2 and 1.5 mm, respectively. Under these conditions the power density (estimated on the basis of the area of laser-target interaction and laser pulse parameters) on the target was $0.8–1.2 \times 10^9$ W/cm^2 and the fluence 3–4.5 J/cm^2, dependent on focusing conditions. The irradiation was performed in two variants: single spot (with a step motor turned-off) and scanned irradiation when the target was moving in front of the beam. Particles released from the irradiated target have been collected by several types of dust catchers: (a) glass plates, adhesive copper foil and carbon stickers placed in the vicinity of the irradiated targets, (b) aluminum cylinders (with a hole for the laser beam) mounted adjacently to the target for collection of larger debris and fine particles released by irradiation (see Fig. 1(B) for details) and (c) copper grids dedicated for studies of fine matter with a transmission electron microscope (TEM) equipped with energy dispersive X-ray spectroscopy (EDX) and diffractometer. In case (a) the dust samples were collected during separate series which consisted of an increasing number of shots (from 10 up to 100) with optimal focusing (i.e. 1.75 mm of the beam semi-major axis) at the treated surface.

Laser-induced desorption (surface heating) has been done using a Nd-YAG laser with pulse length of the order of μs and a power density of up to 800 MW/m^2 at the target; details can be found in [17]. A qualitative analysis of the gas phase composition during laser pulses has been carried out by means of time-of-flight quadrupole mass spectrometry (QMS). The experiments aimed at the comparison of species released from the irradiated co-deposits and from the underlying graphite substrate freed from deuterium. Therefore, the study was performed on two spots on the ALT-II tile, one with the original co-deposit and one previously ablated (at IPPLM) with 50 laser pulses. Desorption has also been done for other types of deuterium-rich carbonaceous layers on graphite.

Irradiated targets and dust generated by pulses have been studied by several techniques to determine the morphology and fuel content. Ion beam analysis (IBA) of dust and deposits formed on catchers was done at the Tandem Accelerator (Uppsala University) by simultaneous nuclear reaction analysis (NRA) and Rutherford backscattering spectroscopy (RBS) using a 2.5 MeV $^3He^+$ beam. The aim was to determine the content of deuterium and carbon by detecting protons released by the nuclear processes: $D(^3He,p)^4He$ and $^{12}C(3He,p)^{14}N$. The nuclear reaction for carbon gives three distinct peaks (p0, p1 and p2) at 5.7, 3.7 and 2.3 MeV respectively, while the nuclear reaction for deuterium gives one single peak at 13 MeV. Because of the background up to 2.5 MeV only the p_0 and p_1 peaks were used for the carbon analysis. Solid-state Ortec Inc. thick (1500 μm) detectors were applied to get the full energy spectrum of the protons. To better resolve the region containing the ^{3}He ions backscattered from the Al substrate of the dust catcher a second solid state thin (150 μm) detector was also used for RBS. The shape of craters formed as result of ablation has been measured by surface profilometry (DEKTAK), whereas TEM, scanning electron (SEM) and optical microscopes have been used for characterizing dust particles collected on different catchers.

3. Results

3.1. Gas phase studies in laser-induced desorption

Mass spectra showing species released during laser-induced desorption from the fuel-rich layer and the underlying graphite substrate are presented in Fig. 2(a and b), respectively, whereas a plot in Fig. 2(c) shows the difference between those two. It is inferred that desorption products consist of HD, D_2 and C_1 and C_2 hydrocarbons (CH_xD_y and $C_2H_xD_y$). The peak at mass 18 is attributed probably both to the release of CH_2D_2 and water adsorbed by the flaking co-deposit when it was exposed to ambient atmosphere before the irradiation. Peaks 20 and 32 result from CD_4 and C_2D_4. The peak at $M = 28$ is associated with carbon monoxide (CO) which is a residual gas in the vacuum system. QMS measurements done in spots where co-deposits have been removed give an evidence and an important confirmation of previous findings [14,18,19] that the bulk of graphite PFC does not retain any significant amount deuterium, i.e. fuel migration into the bulk of graphite is very small.

3.2. Surface and dust studies after laser-induced ablation

Profilometry of the crater formed after 50 laser pulses onto the surface covered by co-deposit has shown that 60 μm thick layer of material was removed. This thickness is somewhat greater (10–20 μm) than that determined for the thickest co-deposits (40–50 μm) on the ALT-II plates [14,16]. The result thus indicates that some substrate material has been removed together with the co-deposit. The ablation generates dust of several different categories.

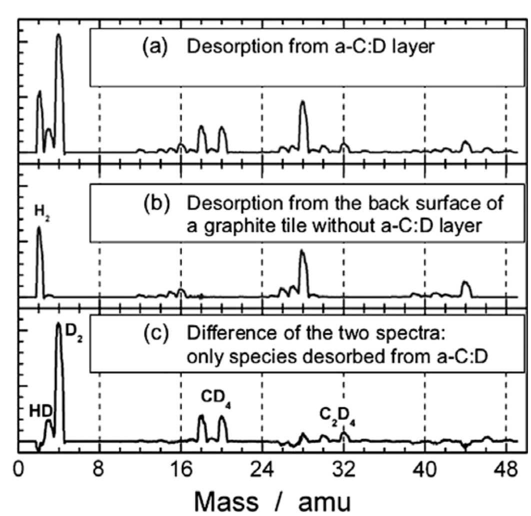

Fig. 2. Mass spectra showing desorption products from co-deposit and graphite substrate.

Dust particles collected on glass plates near the irradiated surface were studied with optical microscopy thus enabling the determination of grain size distribution. The grain size had a broad distribution in range from 2×10^{-3} μm^2 to 1 μm^2, with the mode value of the order of 10^{-1} μm^2. However, the amounts of dust collected on the plates made up only a fraction of all dust generated, because after the experimental session significantly bigger grains and flakes were found in the vacuum chamber. After series of hundreds of shots, the formation brownish layers were observed on metal parts located near the irradiated target. This material was collected with a carbon sticker and the amount of deuterium was determined with NRA. The analysed deposit (3.5–4.1 μm thick) contained about 3.9×10^{17} D cm^{-2} distributed uniformly throughout the layer. Assuming the deposit density of 1.6 g cm^{-2}, the amount of deuterium corresponds to about 1.5 atomic% in carbon matrix, i.e. the D/C concentration ratio is 0.015. This number is 6–8 times lower than that determined on the ALT tiles: D/C concentration ratio of 0.09–0.12 [14,16]. The most important conclusion from the deposit analysis is that not all fuel has been transferred to the gas phase and pumping system. SEM revealed a flaking structure of the layer but its topography was different than that characteristic for stratified co-deposits peeled-off from the ALT plates [14,16].

The application of cylindrical catchers (see Fig. 1(B)) allowed for simultaneous collection and then examination of macroscopic grains (hundreds of micrometers) and ultra-fine matter found on the cylinder walls. Fig. 3 shows the structure of macroscopic dust release from the irradiated target. This dust is very similar to the original stratified flaking co-deposits on ALT [14,16] thus indicating that the laser beam causes mechanical disintegration of co-deposits. The dust contains fuel species. However, the exact comparison to the initial level of deuterium (4.5×10^{19} cm^2 [14,16] in 45 μm deposit) could not be accomplished because of the grain size (300–400 μm) being smaller than the spot of a $^3He^+$ analysing beam. All macroscopic dust was released during the first 5–7 laser shots, but even during following pulses the presence of deuterium was observed with spectroscopy. This suggests that some deuterium-containing species were desorbed from areas adjacent to the beam-ablated spot.

The NRA and RBS examination of fine deposit collected on the catcher walls has been done in several points located 5 mm apart. The deposit contains carbon ($1.3–4.4 \times 10^{17}$ cm^{-2}) and deuterium ($2–9 \times 10^{15}$ cm^{-2}). The corresponding D/C concentration ratio is 0.015–0.02. The film thickness assessed from the RBS measurements is in the range from 200 nm to 650 nm. The structure of such deposit has been determined by TEM. Copper grids were used as collectors of deposits for TEM studies. A bright field image of the deposit and a diffraction pattern are shown in Fig. 4(A) and (B), respectively. They clearly prove that presence of both thin amorphous film and crystalline nano-particles which originate from ablation of the underlying substrate. EDX has found carbon as the only element in the analysed matter. QMS, optical spectroscopy, IBA and TEM studies give very comprehensive and coherent proof that deposits produced in the vicinity of the irradiated target are formed as a result of condensation of hydrocarbons released from laser-treated PFC. This shows that not all volatile products of ablation are pumped out. Therefore, one concludes that laser-induced ablation leads to the re-distribution of fuel to surrounding areas both by producing fuel-rich macroscopic dust from disintegrated co-deposits and by condensation of volatile products.

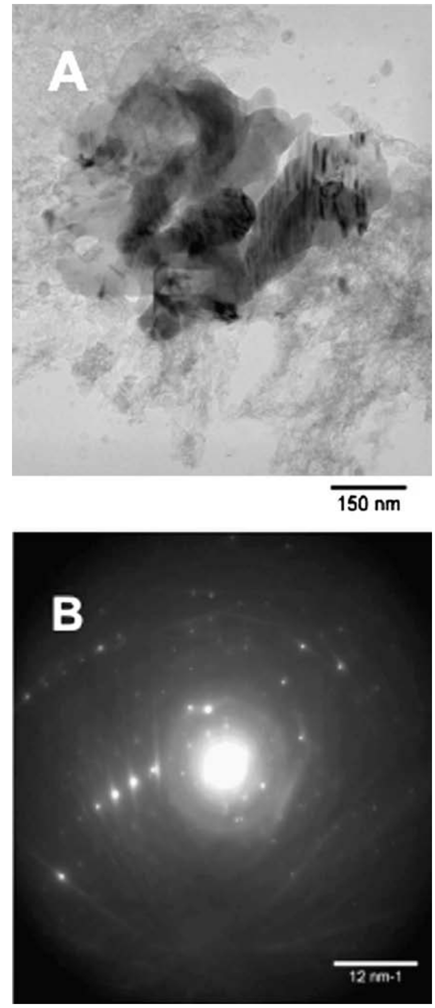

Fig. 4. Bright field TEM image of a conglomerate of amorphous and crystalline particles collected on dust catcher walls (A) and diffraction pattern proving the existence of crystalline matter (B).

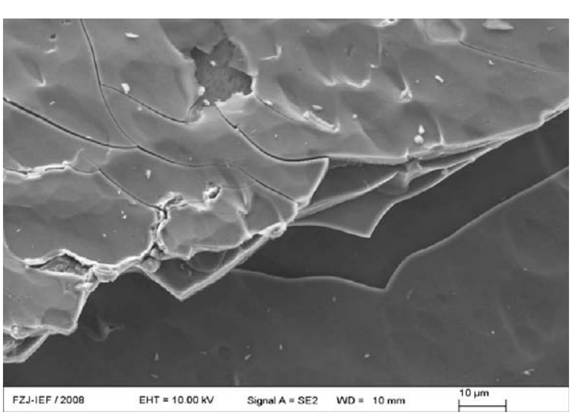

Fig. 3. SEM image of a large dust particle released from co-deposit disintegrated by laser light impact.

4. Concluding remarks and outlook

The study shows that the irradiation of PFC with laser pulses results in the formation of a wide variety of gaseous and solid products, i.e. HD, D_2, C_1, C_2 hydrocarbons and different categories of dust particles. In situ QMS and spectroscopy measurements have proven the transfer of D-containing species to the gas phase. The analysis of fine deposits on catcher walls proves that some deuterated species released from the target condense on adjacent surfaces. A fraction of fuel still remains also in dust particles generated by the laser. Precise fuel balance in all products of co-deposit disintegration is still a challenging task to accomplish, but it becomes evident that photonic cleaning may lead to redistribution of fuel to surrounding areas. Thus dust generation and condensation of desorbed fuel species should be taken into account when developing methods for co-deposit and fuel removal from a reactor-class device. Condensation of hydrocarbons may be an issue if carbon-based wall tiles are applied. One may also suggest that dust production accompanying the cleaning process is necessary regardless of the amounts of fuel which it would still contain, because it is easier to release fuel (tritium) from collected dust than to deal with whole tiles of actively-cooled wall components.

In summary, studies performed with limiter plates from TEXTOR have shown effective removal of deuterium by laser-induced desorption. Co-deposits can also be effectively ablated but – under conditions applied at IPPLM – the process is accompanied by the ablation of some target material. The removal of bulk material will be avoided in the upgraded set-up, where the spectroscopic signal will be used as a feedback.

References

[1] G. Counsell et al., Plasma Phys. Control. Fus. 48 (2006) B189.
[2] W.M. Shu, Y. Kawakubo, M.F. Nishi, Appl. Phys. A76 (2003) 421.
[3] C.H. Skinner et al., J. Nucl. Mater. 313–316 (2003) 496.
[4] A. Semerok et al., J. Appl. Phys. 101 (2007) 084916.
[5] B. Emmoth et al., J. Nucl. Mater. 337–339 (2005) 639.
[6] P. Gąsior et al., Phys. Scr. T123 (2006) 99.
[7] P. Gąsior et al., Czech. J. Phys. 56 (2) (2006) B67.
[8] W.M. Shu et al., J. Nucl. Mater. 313–316 (2003) 584.
[9] C.H. Skinner et al., Phys. Scr. T103 (2003) 34.
[10] F. Le Guern et al., J. Nucl. Mater. 335 (2004) 3.
[11] A. Widdowson et al., J. Nucl. Mater. 363–365 (2007) 341.
[12] C. Grisolia et al., J. Nucl. Mater. 363–365 (2007) 1138.
[13] F. Le Guern et al., Fus. Eng. Des. 81 (2006) 8.
[14] M. Rubel, P. Wienhold, D. Hildebrandt, J. Nucl. Mater. 290–293 (2001) 473.
[15] M. Rubel et al., Phys. Scr. T103 (2003) 20.
[16] M. Rubel et al., Fus. Eng. Des. 81 (2006) 211.
[17] B. Schweer et al., J. Nucl. Mater. 363–365 (2007) 1375.
[18] M. Rubel et al., J. Nucl. Mater. 196–198 (1992) 285.
[19] A. Kreter et al., J. Phys. Conf. Ser. 100 (2008) 062024.