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Plume segregation observed in hydrogen and deuterium containing plasmas produced by laser ablation of carbon fiber tiles from a fusion reactor ☆

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ABSTRACT

The plasma produced by the irradiation of a hydrogen and deuterium containing carbon fiber composite with infrared laser pulses of 4-ns pulse duration has been investigated. The experiments were carried out under argon at reduced pressure. Microscopic analyses of the irradiated sample surface were performed to measure the ablation depth. Time- and space-resolved optical emission spectroscopy was applied to characterize the evolution of spectral line emission as a function of time and distance from the surface. Particular attention was paid to the time-of-flight characteristics of the hydrogen and deuterium Balmer α spectral lines. According to the different atomic masses of both isotopes, the expansion of hydrogen into the low pressure argon atmosphere was found to be slightly faster than that of deuterium. The effect of plume segregation is pressure dependent and tends to increase the analytical signal of heavy atoms with respect to lighter ones during laser-induced breakdown spectroscopy.

1. Introduction

In fusion reactors, plasma — wall interactions due to high particle fluxes result in the erosion of plasma facing components (PFC) leading to the formation of co-deposited layers. This formation is accompanied with fuel deposition (deuterium, tritium) [1]. In order to fulfill safety requirements related to the accumulation of hydrogen isotopes in the vacuum vessel [2], the quantities of trapped deuterium and tritium need to be controlled. Laser-induced breakdown spectroscopy (LIBS), based on the analysis of the optical emission of laser-produced plasma, appears to be an adequate technique to measure the concentrations of hydrogen isotopes in the PFC. LIBS allows for almost non-destructive stand-off and in situ multielemental analysis [3]. However, the technique has one main drawback that consists of the difficulty to perform quantitative measurements. In fact, in case of many complex materials, calibration is inefficient. In that case, the only way to perform quantitative measurements consists to determine the elemental composition of the plasma from the spectra analysis. The so-called calibration-free LIBS measurements [4] generally require the fulfillment of the following conditions: (i) the material ablation process is stoechiometric, (ii) the plasma is in local thermal equilibrium, and (iii) the segregation of elements in the ablation plume is negligible.

In the present study, we have focused our attention to the role of plume segregation due to the mass-dependent expansion velocities of the plasma species. In fact, lighter particles are characterized by larger velocities and their lifetime in the high temperature plasma core is expected to be shorter [5]. As a consequence, the analytical signal may be altered in the way that the concentrations of elements having large atomic mass would be overestimated with respect to the lighter elements. The effect may have an influence on the concentrations measurements of deuterium and tritium trapped in the co-deposited layers on the PFC.

Hydrogen and deuterium have a particular interest to investigate the plume segregation: both isotopes have different atomic masses but almost equal excitation energies. Thus, their relative spectral line intensities are not influenced by the plasma temperature and electron density. The different expansion characteristics of hydrogen and deuterium are therefore easily observable using optical emission spectroscopy [6]. Both isotopes are present in the carbon fiber composite (CFC) tile that was extracted from the fusion reactor Tore Supra for the present study [7].

2. Experiment

Material ablation was produced by an Nd:YAG laser (Quantel, model Brio) delivering pulses of 100-mJ energy and 4-ns duration. The laser was operated at 1064 nm. The laser pulse energy was attenuated to 5 mJ by turning the beam polarization with the aid of a

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half-wave plate and crossing through a polarization analyzer. The laser beam was focused onto the sample surface using a plano-convex lens of 150-mm focal length. According to a spot diameter of 120 µm of the Gaussian beam, a laser fluence of about 50 J cm^{-2} was obtained on the sample surface. The sample was a part of a carbon fiber composite tile of the first wall of the Tore Supra fusion reactor. The tile was extracted several months before the spectral measurements were carried out, and kept in a plastic bag to avoid the contact with humid air. For the experiments, a cuboid of 30×20-mm^2 area and 20-mm height was cut from the tile and placed on a motorized sample holder in a vacuum chamber of 10^{-4}-Pa residual pressure. The chamber was filled with argon at a pressure ranging from 5×10^1 to 5×10^3 Pa during the experiments. The pressure conditions of the experimental chamber were not supposed to affect the loading of hydrogen and deuterium of the sample. The latter was expected to be constant over the time needed to carry out the experiments.

The time- and space-resolved spectroscopic analyses were performed using an imaging spectrometer (Jobin-Yvon, model FHR 1000) of 1-m focal length. The plasma plume was imaged onto the spectrometer entrance slit using two MgF_2 lenses of 200- and 400-mm focal lengths in order to obtain an image magnification of a factor of 2. The two lenses were mounted on motorized translation stages in order to adjust their positions according to the variation of the focal lengths with the observation wavelength. Photon detection at the spectrometer output was ensured using a charge-coupled device (ICCD) matrix detector (Princeton Instruments, model PI-MAX). Using a grating of 1800 grooves mm^{-1}, a spectral resolution of $\lambda/\Delta\lambda\cong2\times10^4$ was obtained. An optical filter was placed at the spectrometer entrance to avoid overlapping with the second observation order of the grating. According to the spectrometer slit width of 100 µm, the emission was captured from a plasma layer of 50-µm thickness. The distance z of the observation zone with respect to the target surface was varied by translating the f = 400 mm lens along the y-axis as shown in Fig. 1.

For LIBS experiments, spatially integrated spectroscopic measurements were performed by imaging the plasma in the direction parallel to the surface normal onto the entrance of an optical fiber of 600-µm diameter. Two lenses of 150- and 37.5-mm focal lengths were used to reduce the image by a factor of 1/4. The captured plasma emission was transmitted via the optical fiber to an Echelle spectrometer (LTB, model Aryelle Butterfly) having a square entrance of 50×50 $µm^2$ dimension. The spectrometer was equipped with an ICCD array (Andor, model iStar 734) to ensure photon detection. The spectral resolution was of about 1×10^4.

Synchronization between laser pulse and ICCD gates was ensured using the Q-switch pulse trigger output of the Nd:YAG laser. To enhance the signal-to-noise ratio, each recording was performed by accumulating the signal over several ablation events. Therefore, different sites were irradiated by applying one or several laser pulses on each site. The sites were separated by a distance of 300 µm.

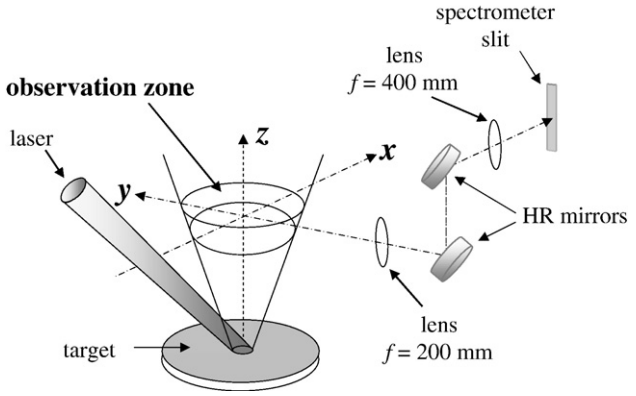

Fig. 1. Observation geometry for spectral imaging. The plume's symmetry axis (z-axis) is perpendicularly to the surface. A plasma layer of 50-µm thickness is selected for observation by imaging the plasma on the entrance slit of the spectrometer.

Table 1
Ablation depth produced by a single laser pulse as a function of argon pressure. The measurement uncertainty is estimated to about 20%.

Argon pressure (Pa)	Ablation depth (µm)
5×10^1	≈4
5×10^2	≈2
5×10^3	≈0.5

3. Results and discussions

3.1. Ablation depth measurements

The evaluation of the relative abundance of deuterium and hydrogen as a function of depth in the bulk of the CFC tile is of crucial interest for the present study. It requires the knowledge of the ablated depth produced by a single laser pulse. We have thus deduced the ablation depth from microscopic observations of the laser-produced craters. To increase the measurement precision, ten craters were produced for each condition. Each crater was obtained by applying 5 successive laser pulses, assuming a linear increase of crater depth with the number of applied laser pulses [8]. We observe that the ablation depth strongly decreases with increasing argon pressure (see Table 1). This behavior is attributed to the plasma screening effect that is particularly efficient for infrared radiation [9]. Because of the plume confinement at larger pressure, the increased collision rates lead to stronger absorption via inverse bremsstrahlung [10]. As a consequence, the energy coupling between the incident radiation and the material is lowered, and the volume of ablated matter is reduced.

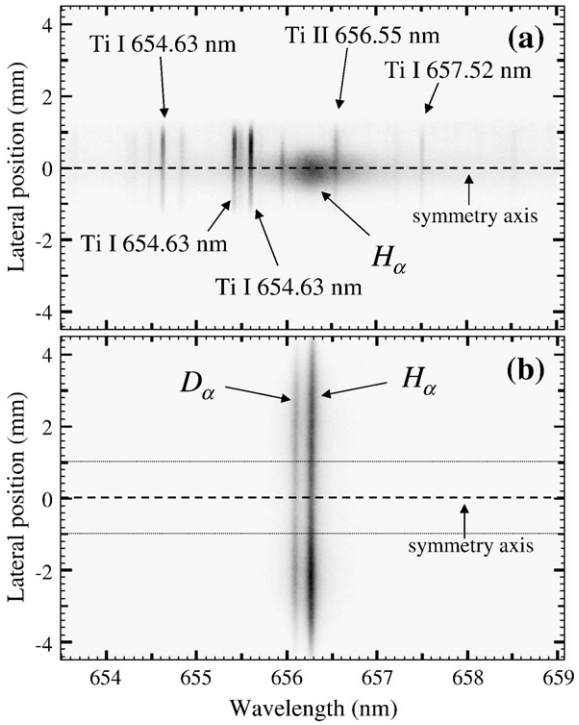

Fig. 2. Spectral images recorded during ablation of titanium in ambient air (a) and the CFC sample argon at 5×10^2-Pa pressure (b). The recordings were performed with a delay and an observation gate duration of 1 and 2 µs, respectively. The observation distances from the target were 0.75 mm (a) and 1.0 mm (b). The lateral position corresponds to the x-axis of Fig. 1.