Vacuum laser-produced plasma for analytical application in fusion technologies

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Abstract. LIBS is a well established tool for qualitative, semi-quantitative and quantitative analysis of surfaces, with micro-destructive characteristics and some capabilities for stratigraphy. In this work, the depth profiling capabilities of LIBS has been checked by determining the composition of multilayered samples simulating the plasma facing components fusion device covered with co-deposited impurity layers. A new experimental setup has been designed and realized in order to optimize the characteristics of a LIBS system working at low pressure and remotely.

1. Introduction

High temperature plasmas of hydrogen isotopes are peculiar of thermonuclear fusion devices. The study of plasma-wall interaction [1-3] is of paramount importance for avoiding both damage of plasma facing components (PFCs) and pollution of the plasma. PFCs are eroded and the material is deposited by the edge plasma in different areas of the tokamak. So the composition of PFCs’ surface changes strongly from the original. To ensure safety operation a regular monitoring of the tritium inventory and the deposited material has to be applied. To this respect Laser-Induced-Breakdown-Spectroscopy (LIBS) [4-5] may be suitable for in-situ characterization of the conditions of PFCs.

The characterization of first wall surfaces by LIBS, which means the local measurement of thickness and composition of deposited layers including tritium inventory, could be carried out by probing with the laser beam different wall areas between tokamak discharges. Depending on the power density and pulse duration of the laser radiation, wall particles are released by ablation and are spectroscopically analysed. The spectral emission of such LIBS plasma is governed by atomic physics processes involving collisional excitation and decay rates of several states of the neutrals and ions in the plume. For optically thin plasma the line intensity for transition between the energy states \(k\) and \(i\) can be given by [5]

\[
I_\alpha = FC_\alpha g_k A_{k,i} e^{-E_k/k_B T} \frac{U_\alpha(T)}{U_\alpha(T)}
\]  

(1)
where $g_d$ is the state degeneracy and $A_d$ the Einstein coefficient for spontaneous emission, $T$ is the plasma temperature, $U_d(T)$ is the partition function, $K_B$ is the Boltzmann constant and $F$ is a constant to account for particular experimental conditions (optical arrangement and photon counting efficiencies) and plasma density (depending on material and absorbed laser energy).

As an analytical technique, LIBS has demonstrated its unique versatility [5-7], allowing fast contact-less analysis of almost any type of material, from solids to liquids and gases also examined at low pressure [8], each time allowing to adapt the technique to the spectral requirements of diverse practical analytical problems. The spectroscopy of the radiation emitted by the Laser Induced Plasma (LIP) may be used to obtain the characteristic physical parameters, such as the temperature, the electron number density, the atom and ion number densities, and the relative concentration of different element present in the target.

In the framework of the European Fusion Development Agreement (EFDA) task WP11-ETS_DTM-01 “Dust and Tritium Management” the objective of this work is to advance and qualify the operation of LIBS on a fusion device (tokamak) by performing Deuterium inventory in targets specifically prepared to simulate PFCs covered with impurity layers codeposited with fuel. The optimization criteria here considered are the capability of a LIBS system in working at low pressure and remotely, at a distance of some meters from the target. The optical system used is sketched in Figure 1.

![Figure 1. sketch of the LIBS apparatus.](image)

The apparatus is composed of a vacuum chamber, 35 cm diameter in which the sample is placed vertically on a two axis motorized translation stage remotely controlled for changing the measurement point. LIPs were obtained by using a Q-switched Nd:YAG laser (model handy, Quanta System) emitting at the fundamental wavelength of 1064 nm. Each laser pulse has a duration of 8 ns, while repetition rate is 10 Hz. The laser beam was focused onto the sample by a 200 cm focal length quartz lens through a 150 cm length vacuum arm closed at the end by an optical window highly transparent to infrared radiation. With an average power of 140 mJ and a spot diameter of 1.3 mm on the sample surface the energy fluence released was about 11 J/cm² per single shot.

To realize remote plasma light detection and simulate the dimensional constraints found in a real tokamak a plane mirror with a 10 mm hole in the center was placed in front of the focusing lens and aligned with the laser beam (passing trough the hole). In this way the plasma light exiting from the optical window, collinear with the laser beam, is reflected by the mirror at an angle of 45° respect to the laser beam and is collected at a distance of 200 cm from the sample surface.
All the measurements were carried out at a residual pressure of \(1-3 \times 10^{-5}\) mbar. The collection optics is composed by a 15-cm focal length fused quartz lens, 50 mm aperture. The lens focuses the plasma light into an optical fiber bundle, composed of 37 fibers, 100 µm core, f# 4, connected to a 100 µm entrance slit of a TRIAX 550 ISA JOBIN-IVON spectrograph, f# 6.4. For a diffractive grating of 2400 g/mm the wavelength resolution at 500 nm is around 0.1 Å. Spectra were recorded using a gated ICCD (DH532-18F, Andor), whose gate aperture was synchronized with the laser burst by mean of an electronic trigger.

1.1 Samples
A set of diamond-like carbon (DLC) and mixed W:Al:C (with nominal concentrations W(5-10%):Al(45-48%):C(45-48%)) layers were codeposited with D on W substrates by vacuum arc deposition method.

The mixed sample simulates ITER-like PFCs surface composition in the redeposition zones [9]. Al was used as proxy for Be (not used in these samples due to its toxicity) that, together with W and C will be one of the principal constituent of ITER PFCs [9]. The nominal thickness of the superficial layer was ≥ 3 µm. The samples, round shaped, with a diameter of 25 mm, were first characterized with Thermal Desorption Spectroscopy (TDS) [10] and Nuclear Reaction Analysis (NRA) [12] to determine the concentration of each element.

2. Results

2.1. Depth profiling
To highlight the depth profiling capabilities of LIBS we have repeatedly hit the same point of the sample with subsequent laser shots ablating the material coming from deeper and deeper layers of the sample and recording at the same time the evolution of the spectral signal as a function of the shot number.

To do this the spectral region between 425 and 445 nm has been selected because in this range it is possible to find intense spectral emissions of both C (in particular CII at 426.73 nm) and W. Figure 2 shows the sequence of spectra obtained for successive laser shots on the sample DLC; it is possible to clearly recognize the evolution of the spectral signal that shows the intense spectral emission of the CII up to about the 21° laser shot where subsequently emerges a very different spectral track, ascribable to the W substrate, to indicate that the surface layer of codeposited carbon was completely ablated.

![Figure 2. Sequence of spectra in the spectral region between 425 and 445 nm.](image-url)
Similar results were also obtained on the mixed sample W: Al: C, not only in the spectral region above but also in the spectral region between 392 and 405 nm, where strong spectral emissions attributable to tungsten and aluminum are detectable.

The obtained results suggest that monitoring the intensity of the spectral lines of each species of interest (e.g. Al I at 394.4 and 396.2 nm and W I at 400.9 nm for the substrate) allows to control its concentration in the layer under investigation. Figure 3 shows a graph of some spectral emissions of C and W in the spectral range of figure 2 as a function of laser shot number. The transition from the mixed layer to the bulk material is clearly seen around the laser shots # 20-25.

Considering a depth of the surface layer of 3 µm (as shown by the NRA measurements) an ablation rate of 0.13 µm/laser pulse is obtained.

Figure 3. LIBS intensity as a function of laser shot for some of the C and W atomic and ionic lines in the spectral region 425-445 nm.

This value depends on many factors including the laser power, the profile of the beam, the focusing optics, the duration of the laser shot [13-14], and can not be regarded as an absolute reference.

However is worth remembering that a further increase in depth profiling resolution could be obtained simply decreasing the laser fluence (provided that the ablation threshold is still reached and the emission signals emerge from the background noise), while a greater laser power could provide a higher ablation rate and a higher spectral signal at least until effects of saturation of the spectral lines, mainly due to reabsorption of the laser energy by the same plasma are not observed [15], limiting the interaction with the target and the ablated mass per single shot.

2.2. Detection of trace elements.

The evaluation of the relative and absolute abundance of deuterium as a function of depth in the bulk of the tiles is of principal interest for the present study.

It has been possible to detect the D$_a$ spectral line at 656.1 nm, well distinguished from H$_a$ spectral line at 656.29 [16]. The graph of Figure 4a shows the LIBS signal in the spectral region of interest (between 653 and 659 nm) on sample DLC. H$_a$ and D$_a$ emissions are clearly visible and well resolved, together with the spectral emissions of two ionic C lines at 657.8 nm and 658. 29 nm respectively.

Through the preliminary NRA and TDS measurements we have mentioned before, the atomic concentration of D in the codeposited surface layer was estimated to be around 3%, a value in agreement with what obtained in the present LIBS measurements once the calibration free method was applied [17]. In the graph of Figure 4b the evolution of the spectral emission as a function of laser
shots is reported, it shows a reduction of the spectral signal around shot 20-25, consistently with the evolution observed between 425 nm and 445 nm.

Figure 4a. LIBS spectrum of sample DLC in the region between 654 and 660 nm, the spectral emission of hydrogen (H$_2$) and deuterium (D$_2$) is shown, in addition with the emission of two C ionic lines

Figure 4b. Evolution of the LIBS signal as a function of laser shot in the same spectral region of Figure 4a. As in Figure 2 the red spectrum was acquired at the 21$^{th}$ laser shot

3. Conclusions

We set up a LIBS apparatus able to excite and detect plasma emission at long distances and in vacuum.

The targets used were prepared in order to reproduce the surface layers of PFCs of current and new generation fusion devices.

It was possible to reveal both the atomic composition of the targets surface coating and the presence of deuterium in trace amounts on the surface layer, as confirmed by independent measurements.

From this study emerges that the LIBS technique is suitable to operate as stand-off diagnostics, both for monitoring the PFCs erosion and the fuel deposition on the fusion device wall.

Further developments of this study aim to determine the relative concentrations of the detected elements and the influence of a toroidal magnetic fields (<3 T) on the plasma plume shape and performance, testing the apparatus in a real tokamak like FTU (Frascati Tokamak Upgrade).

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