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Application of laser-induced breakdown spectroscopy for characterization of material deposits and tritium retention in fusion devices

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HIGHLIGHTS

- This study provides an *in situ* method to monitor the depth profile of co-deposition.
- It also proposes an optimized experiment conditions for laser removal.
- Fuel retention/removal mechanisms have been proposed.
- Quantitative methods for *in situ* monitor the amount of the deposition composition are proposed and compared with other methods.

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ABSTRACT

Laser-induced breakdown spectroscopy (LIBS) is discussed as a possible method to characterize the composition, tritium retention and amount of material deposits on the first wall of fusion devices. The principle of the technique is the ablation of the co-deposited layer by a laser pulse with P (power density) $\geq 0.5\,GW/cm^2$ and the spectroscopic analysis of the light emitted by the laser induced plasma. The typical spatial extension of the laser plasma plume is in the order of 1 cm with typical plasma parameters of $n_e \approx 3 \times 10^{22}\,m^{-3}$ and $T_e \approx 1$–$2\,eV$ averaged over the plasma lifetime which is below 1 μs. In this study "ITER-Like" mixed deposits with a thickness of about 2 μm and consisting of a mixture of W/Al/C and D on bulk tungsten substrates have been analyzed by LIBS to measure the composition and hydrogen isotopes content at different laser energies, ranging from about $2\,J/cm^2$ ($0.3\,GW/cm^2$) to about $17\,J/cm^2$ ($2.4\,GW/cm^2$) for 7 ns laser pulses. It is found that the laser energies above about $7\,J/cm^2$ ($1\,GW/cm^2$) are needed to achieve the full removal of the deposit layer and identify a clear interface between the deposit and the bulk tungsten substrate by applying 15–20 laser pulses while hydrogen isotopes decrease strongly after the first laser pulse. Under these conditions, the evolution of the spectral line intensities of W/Al/C/hydrogen can be used to evaluate the layer composition.

1. Introduction

Analysis and understanding of the wall erosion, material deposition and the associated tritium retention by co-deposition with eroded wall material is of high importance for a fusion reactor operation and, in particular, to fulfil safety requirements for tritium retention [1–4]. Laser based technique like laser-induced desorption spectroscopy (LIDS), laser-induced ablation spectroscopy (LIAS) and laser-induced breakdown spectroscopy (LIBS) are under development by various groups [1–7] to analyze *in situ* the fuel retention and the material deposition. The LIBS method is under investigation both in the laboratory experiments (FZJ and DUT) and *in situ* in the TEXTOR tokamak in Forschungszentrum Jülich (FZJ) together other EU fusion labs. The aim is to study its feasibility for quantitative wall analysis under the special constraints of fusion environments, such as high vacuum, large distances of the LIBS-plume to the detector, presence of the magnetic field and unknown physical properties of the deposited layers. LIBS should provide not only the localization of the fuel retention and the qualitative composition of the layer, which can be used to discriminate

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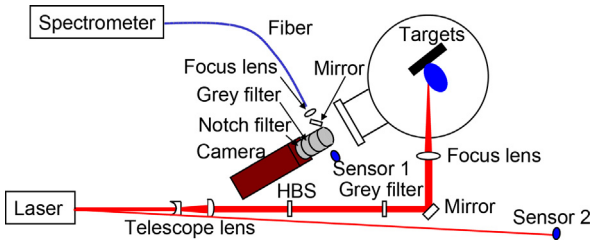

Fig. 1. Experimental setup of the laser induced breakdown spectroscopy in Lab.

the co-deposited layer from the substrate and, possible, to monitor removal processes, but also the quantitative composition of the co-deposited layer and its fuel retention. The applicability of LIBS for the depth profiling and determination of the composition of multilayer structures [8] and mixed material layers [9,10] has been verified. Under conventional conditions, quantitative analysis of LIBS relies on the carefully prepared reference samples with the composition similar to the composition of the analyzed sample, which is difficult to create for the co-deposited layers with in advance unknown composition under real tokamak conditions. The so-called "calibration-free" method provides another way to obtain the elemental composition [11]. However, this method relies on the knowledge of the plasma parameters (n_e and T_e) which are strongly time dependent during the LIBS plasma and also on the accuracy of the atomic database. Thus the optimization of the absolute calibration of LIBS under fusion conditions remains an important issue for further R&D. This is also the main topic of this paper.

2. Experimental set-up

The experimental set-up for the laser-induced breakdown spectroscopy in the laboratory is shown in Fig. 1. In this study "ITER-Like" mixed deposits with a thickness of about 2 μm consisting of a mixture of W/Al/C with some amount of deuterium on bulk tungsten substrates have been produced by plasma (CVD) and arc plasma deposition technique (DIARC® [12]) and magnetron sputtering, Al was chosen as a replacement for Be [13–15]. The layers have been characterized by surface analysis beforehand (NRA, RBS, TDS and SIMS) and then analyzed by LIBS for their composition and hydrogen content at different laser energies, ranging from about 2 J/cm^2 to about 17 J/cm^2 for 7 ns laser pulses on a typical spot area of 3.67 mm^2. The corresponding power densities, which are absorbed by the target, depend on the reflectivity properties and surface roughness of the sample. The samples were mounted vertically on an XYZ translation stage to change the position of the spot on the sample when it is necessary. A Nd:YAG laser beam at fundamental wavelength 1064 nm has been focused onto the target with an incidence angle of about 45°. A telescope lens was applied to expand the diameter of the beam the energy is monitored *in situ*. After reflection by a mirror, the expanded beam was focused by a lens with a focal length of 70 cm onto the sample. The laser beam caustic is adjusted inside the sample to avoid air breakdown in front of the sample. The laser-induced plasma emission light is collected in a direction parallel to the target surface by a fibre which is connected to a two-dimensional camera and a cross-dispersion Echelle spectrometer (resolution $\lambda/\Delta\lambda \approx 20{,}000$) with a wavlength range from 365 nm to 715 nm equipped with a two dimensional CCD camera. The measurements are performed in a vacuum chamber which is evacuated up to 10^{-7} mbar by a turbomolecular pump system. Two fast optical sensors (time resolution ~1 ns) and a fast CCD camera with band pass filter and notch filter in front are used to monitor *in situ* the laser pulse and the plasma radiation, respectively.

3. Results and discussion

3.1. Composition and depth profile analysis

At the beginning, the elemental analysis of the ITER like deposits was obtained by the LIBS analysis which would also be done at the first for the *in situ* analysis of the first wall in fusion devices. The spectra show the composition to be Al, C and W, with some contribution of hydrogen and deuterium. Fig. 2 shows the emission lines after 1, 14 and 50 pulses under the energy density of 17 J/cm^2. From the spectra, we see that the spectral lines of Al and C have largely decreased while the W lines have increased, showing that the layer almost removed and the W substrate reached. Some small Al and C signals are still detectable after 50 pulses, which is due to the removal of material from the crater border although the spot centre is completely ablated.

At the same time, the hydrogen isotopes in the co-deposited layer have been detected by LIBS spectra. Fig. 3 presents some H_α and D_α spectra for lasers pulses of 1, 10 and 50 with the energy density of 17 J/cm^2 and the whole evolution of the H_α, D_α integrated intensities for different laser energy densities. The H_α, D_α line intensities decrease significantly after the first pulse for all energy densities. The first H_α signal decreases with the energy density whereas the first D_α signal stays a constant. The origin of the hydrogen signal is mainly from surface absorption, but also from inside the deposit due to its porosity, from the water while the deuterium signal originates from desorption and ablation and its concentration is nearly constant. At the first several laser pulses, the H_α, D_α intensities stay at some lower level under high energy densities, while they are almost removed by a single laser pulse with lower energy densities. The data indicate that a great part of the stored deuterium can be released thermally in a single laser pulse. However, the small amount hydrogen and deuterium released by ablation can only be detected under high energy density in which the produced plasma absorbs a significant part of the incoming laser energy since the energy for ablation are saturated above the threshold [6]. One may also speculate that the remained fuels are formed during the first laser spot heating, by agglomeration of fuel on deep trap sites, such as bubbles.

LIBS can convincingly provide a depth profile of the compositions in the deposition layer via pulse to pulse ablation. Fig. 4 shows the behaviour of emission lines of Al, W and C for several energy densities as a function of the laser pulse. The depth profiles

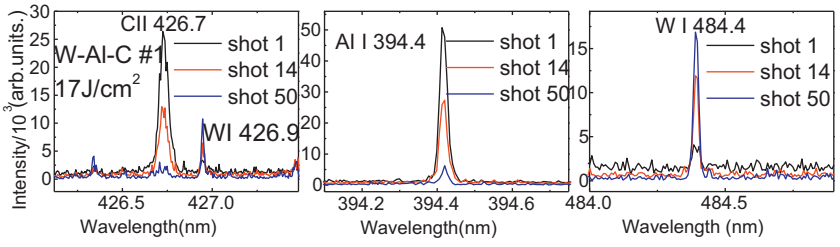

Fig. 2. Typical Al, C and W lines at pulses 1, 14 and 50 in the same laser ablation spot.