Optimisation of LIBS parameters for analyzing co-deposited layers in ITER

J. Karhunen[1], A. Hakola[1], J. Likonen[1], A. Lissovski[2], P. Paris[2], M. Laan[2], C. Porosnicu[3], C.P. Lungu[3], K. Sugiyama[4] and JET EFDA Contributors*

[1] *VTT Technical Research Centre of Finland, Association Euratom-Tekes, 02044 VTT Finland*
[2] *Institute of Physics, University of Tartu, 51010 Tartu, Estonia*
[3] *INFLPR, MEdC EURATOM Association Magurele-Bucharest, 077125, Romania*
[4] *MPI für Plasmaphysik, EURATOM Association, D-85748 Garching, Germany*
**See the Appendix of F. Romanelli et al., Proceedings of the 24th IAEA Fusion Energy Conference 2012, San Diego, US*

Introduction

Laser-induced breakdown spectroscopy (LIBS), based on studying the spectrum emitted in a laser ablation process, is a potential method for *in situ* studies of the compositions of mixed layers deposited on the first walls of future fusion reactors [1]. By LIBS, these layers could be monitored in between plasma discharges without the need for removing any wall components.

In this contribution, the aim is to find optimal experimental parameters for studying ITER-relevant materials, especially D-containing mixed layers of Be and W. This is done by varying the fluence of the laser beam, triggering of the measurements, and the laser wavelength.

Experiments

In the experimental set-up used in these studies, the sample is placed in a vacuum chamber with base pressure below 10^{-6} mbar. Ablation is induced by a pulsed Nd:YAG laser providing 5-ns pulses at 1064 nm with a maximum energy of 650 mJ. The laser beam is focused on the sample by a 500-mm lens, and the emission is detected perpendicular to the laser beam by an Andor SR-750 spectrometer equipped with an Andor iStar 340T ICCD camera.

A set of 500–2500-nm coatings with different Be-W atomic ratios, ranging from 100:0 to 75:25, was produced at MEdC by the TVA method [2] to simulate ITER-relevant mixed layers. To study fuel retention, some samples were implanted with 200-eV D atoms. In addition, experiments were made using samples drilled from the inner divertor tiles of ASDEX Upgrade and JET to investigate real W- and Be-containing tokamak samples with larger D contents than in the test coatings. The ASDEX Upgrade tiles had 0.5–1.5-μm thick W coatings on graphite and were exposed to plasma operations during its 2009 campaign. The JET tiles were pure CFC and were exposed during the period 2007-2009 having up to 70 μm thick Be-, C-, and D-rich co-deposited layers on their surfaces; Be originates from regularly performed evaporations.

Optimisation of the laser fluence

Fluences within 1–38 $\frac{\mathrm{J}}{\mathrm{cm}^2}$ were achievable in the experiments. The effect of the fluence on the detected signal and the depth resolution of the LIBS studies was investigated by studying the intensity levels and widths of atomic and ionic spectral lines of Be and the ablation rate of the studied layer as functions of the fluence. All these parameters increase approximately linearly with the fluence. This can be seen in Figure 1a, where these quantities are normalized to their maximum values. The increase is the steepest for the intensities of Be(I) and Be(II) lines, whereas the respective line widths show a relatively weak fluence dependency.

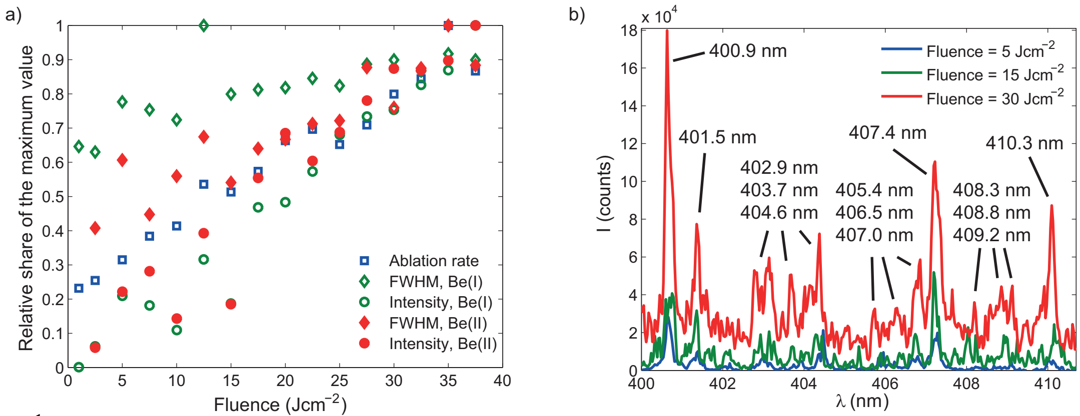

Figure 1: a) The behavior of the intensities and widths of Be(I) and Be(II) lines and the ablation rate of the studied coating as a function of the laser fluence, b) W(I) lines measured with different fluences, showing the increasing quasi background due to the weak W(I) lines around the most intense lines at 400.9 nm, 407.4 nm, and 410.3 nm.

In the optimal case, the observed spectral lines are intense and narrow, and the ablation rate is low to keep the depth resolution of the measurements acceptable. The ablation rate depends heavily on the structure of the layer and was observed to range from 0.06 $\frac{\mu m}{shot}$ to 15 $\frac{\mu m}{shot}$ between different samples at a fixed fluence. Thus, the main attention was paid to the intensity and line width. Considering the rapid increase of intensity in Figure 1a compared to that of the line width, the optimal fluence was estimated to be rather high at 20–25 $\frac{J}{cm^2}$.

The signal-to-noise ratio of the most intense Be lines is acceptable for all fluences. In the case of W, the most intense lines are surrounded by many weaker lines, as shown in Figure 1b, so that the quasi background they create increases with fluence together with the lines used for diagnostics. Thus, the signal-to-noise ratio of the W lines remains approximately constant as the fluence increases, but the generally low intensity level of the W lines favours high fluences.

In the implanted samples, D was not detected due to its low concentration in the samples, but a clear D signal was recorded with several successive laser pulses from the JET and ASDEX Upgrade samples. As can be seen in Figure 2a, higher fluences provide more intense D_α lines, which substantially increases the signal-to-noise ratio. High fluences are also supported by Figure 2b, where the depth profiles of D at different fluences are shown: the curves remain constant independent of the fluence. Thereby, the detection of D is not disturbed by too prompt removal.

Temporal adjustment of the measurements

The triggering scheme of the measurement system was mastered by the laser which sent the initial measuring command to the ICCD camera 500 ns before the release of the Q switch.

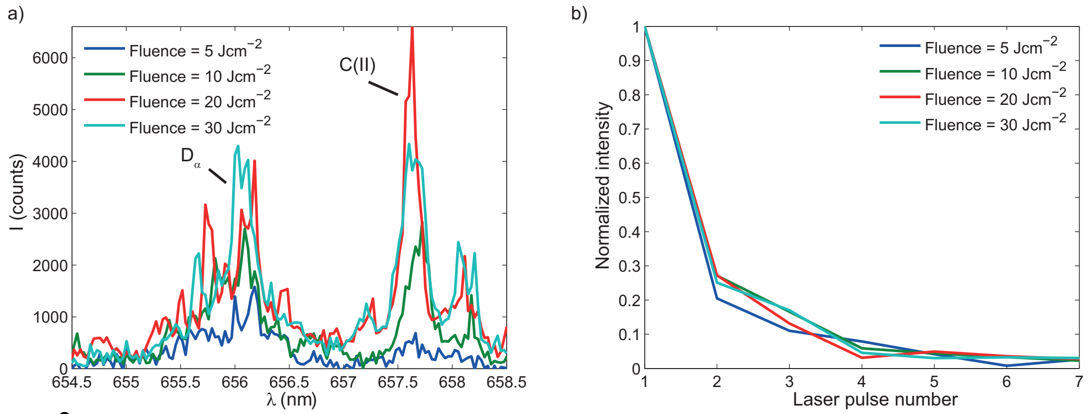

Figure 2: a) The signal-to-noise ratio of the D_α line increases with fluence, b) The shape of the depth profile of D remains constant with increasing fluence.

The delay between this triggering signal and the onset of recording was adjusted by a pulse generator, although it has to be noted that the real delay is somewhat longer due to an internal delay of 30–35 ns of the measuring devices. The width of the measuring gate was 500 ns.

For detecting Be and W in the coated samples, 550 ns was found to be the optimal delay: the background continuum had faded away but both atomic and ionic lines were still detected. However, in the JET samples ionic C lines were observed in the close proximity of the Be(II) line at 467 nm and the D_α line with shorter delays around 450–500 ns. In case ITER will begin its operation with C in the divertor, shorter delays could thus be used for studying co-deposition of Be and D with C. For the detection of Be or D only, longer delays provide C-free signal with a low background level.

Comparison between laser wavelengths: 1064 nm vs. 266 nm

A set of experiments was also performed by using the fourth harmonic, 266 nm, of the laser wavelength to investigate the effect of wavelength on ablation. After the harmonic generation, the pulse length was shortened to 4 ns and the pulse energy restricted to 120 mJ, so that the fluence was reduced to approximately 6 $\frac{\mathrm{J}}{\mathrm{cm}^2}$. Thus, the results were compared to those obtained by IR pulses with a fluence of 5 $\frac{\mathrm{J}}{\mathrm{cm}^2}$.

The intensity of the beryllium lines was observed to be 1–2 orders of magnitude lower than using an IR laser. This is most probably due to the inefficient absorption of the energy of the laser pulse into the LIBS plasma. The emission was also shorter than that induced by IR pulses such that the measurement delay had to be slightly shortened and could not be adjusted as freely as in the IR regime.

In the UV regime, the signal was also found to be much more heavily dependent on the studied layer than in the IR case: for pure Be coatings and the JET samples, the lines were very weak and ionic emission barely detectable, whereas for mixed coatings with Be-W atomic ratios of 75:25 and 85:15, much stronger signals for both Be(I) and Be(II) lines were observed. This might suggest that the fluence was insufficient to induce good emission with strongly crystalline pure Be and the low Be content in the JET samples, but ablation was more efficient for the Be rich and less crystalline Be-W mixtures. The W lines were extremely weak for both the mixed coatings and the ASDEX Upgrade samples, and only traces of D were detected in the JET samples. These are mostly due to the generally low intensity level of the UV experiments: at higher fluences, UV pulses have been observed to induce a better signal-to-noise ratio for the W lines than IR pulses [3].

Due to the much smaller interaction volume of the UV pulses with the sample surface, profilometer studies revealed a 10-fold decrease in the width of the ablation craters compared to IR experiments. This indicates that only the central part of the less energetic UV beam ablates the surface and the widely spreading heating effect plays an important role in IR-induced ablation. The smaller craters enable denser sampling, if UV laser is used in LIBS experiments.

Conclusions

Several mixed coatings of Be and W as well as samples from ASDEX Upgrade and JET were studied by LIBS with different fluences, delays, and laser wavelengths. The results show that the best signal for Be, W, and D is observed at high fluences around 20–25 $\frac{J}{cm^2}$, which, however, sets limits for the depth resolution of the LIBS studies. By adjusting the measuring delay, Be and D could be studied simultaneously with C, enabling feasible investigation of co-deposited layers. When an UV laser wavelength at a low fluence was used, the detected signal decreased substantially, and the detection of materials with low concentrations became complicated. Hence, higher fluences seem to be required with an UV laser than in the IR case, which can degrade the depth resolution when studying the easily ablated co-deposited layers. Furthermore, a UV-compatible measuring system is more difficult to implement in a tokamak environment than is the case with IR.

Due to the extremely different ablation properties of specifically fabricated coatings and deposited layers on actual tokamak samples, the future tokamak-relevant LIBS studies will concentrate on the latter ones. Moreover, the optical set-up will be renewed to provide higher fluences also in UV operation by reducing the laser spot size.

This work was supported by EURATOM and carried out within the framework of the European Fusion Development Agreement. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

References

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