



# Laser-induced breakdown spectroscopic characterization of impurity deposition on the first wall of a magnetic confined fusion device: Experimental Advanced Superconducting Tokamak

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## ABSTRACT

Our recent investigations have indicated the potential of laser-induced breakdown spectroscopy (LIBS) for analysis of the co-deposited layers on the first wall in the Experimental Advanced Superconducting Tokamak (EAST) in the vacuum environment. Detailed information of compositions at the superficial and in-depth positions of the first wall of divertor tiles can be obtained by analyzing the spectra from 200 to 980 nm. The decrease in concentrations of the depositional elements (such as D, Li) was clearly observed in the depth from 0 to 100  $\mu\text{m}$ , but the concentrations of the substrate elements were found to be relatively uniform in the depth after dozens of laser pulses. The linear correlation approach has been applied for improving the impurity depth profile accuracy and identifying the interface boundary between the deposition layer and the substrate for the first time. This would help us to develop LIBS technique to monitor the fuel retention and impurity deposition on the first wall of EAST.

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## 1. Introduction

EAST is a fully superconducting tokamak with advanced divertor configurations [1]. In an operating tokamak chamber, the first wall and the divertor tiles must withstand harsh radiation, intense particle bombardment, and high heat loads. Depending on their location, the plasma facing components (PFCs) can be primarily influenced by erosion and deposition [2,3]. In deposition-dominated zones, these impurities would transport into the central plasma and affect the operation performance by radiation. An effective in-situ method of monitoring elemental impurity deposition and fuel retention in PFCs is desired.

Laser-induced breakdown spectrometry (LIBS) is a straightforward in situ tool for three-dimensional chemical analysis of samples on the micrometric scale without any pre-treatment in fusion environment [4–10]. In addition, with the help of data about ablation rate of materials, we can use LIBS spectra recorded with different numbers of laser shots to determine depth profiles of the different elements in PFCs. The present investigations are designed to check the depth profiling capability of the LIBS technique as an in-situ diagnostic method for the determination of deposition layer on PFCs.

## 2. Experimental

### 2.1. Laboratory set-up

A schematic view of the experimental set-up of LIBS in our laboratory is shown in Fig. 1. Samples were mounted on an XYZ translation stage inside a vacuum chamber which was equipped with quartz windows. The vacuum chamber was pumped down to a pressure of  $6 \times 10^{-6}$  mbar to simulate the pressure condition in the fusion vessel. Ablation was produced by a Q-switched Nd:YAG laser (Brilliant Eazy, Quantel) with 5 ns pulse width and 10 Hz repetition operating at the fundamental wavelength of 1064 nm. The laser beam was focused on the sample surface by a lens with a focal length of 250 mm. The maximum pulse energy was 300 mJ. The plasma light emission was collected in a direction perpendicular to the target surface by collimating and focusing lenses with focal lengths of 100 mm and 10 mm respectively. These lenses guided the light into an optical fiber bundle coupled to a spectrometer. A spectrometer, LIBS2500 + (Ocean Optics Inc., US) detection system with seven linear silicon CCD array detectors, made it possible to analyze the plasma emission in wavelength range from 200 to 980 nm with the optical resolution of 0.1 nm at a single laser pulse. The current optical collecting system was chosen to adapt our experimental condition, which could be modified easily (with suitable collecting lens) for the in-situ diagnosis of the PFCs of EAST by LIBS technique.

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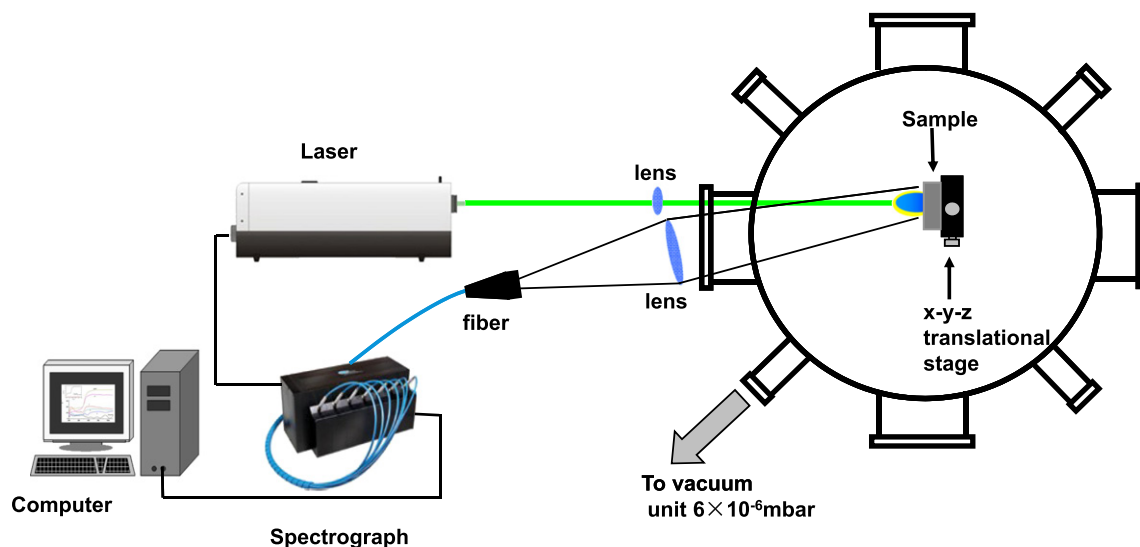


Fig. 1. Schematic view of the experimental setup in Lab.

## 2.2. Samples

During the 2010 and 2012 campaigns, the divertor tiles of EAST were uniform and were made of multi-element doped graphite GBST1308 (1% B4C, 2.5% Si, and 7.5% Ti) with about 100  $\mu\text{m}$  thick SiC gradient coatings [11]. The tiles were used to cover all of the divertor panels and were designed to endure high heat flux without large erosion or damage. Five typical samples were prepared for our LIBS experiments. The first one was a piece of original graphite tile (not exposed to plasma). The second one (named DM2-2010) and the third (named ID5-2010) were installed on the inner target plate and on the dome of the upper divertor, respectively. They were exposed to plasma discharges during the 2010 campaign. The fourth one (named DM2-2012) and the fifth (named ID5-2012) were installed on the inner target plate and on the dome of the upper divertor, respectively. Both of them were exposed to plasma discharges during 2012 campaign.

## 3. Results and discussion

### 3.1. Surface morphology and composition

Details of surface features of the divertor tiles were examined by scanning electron microscope (SEM). Fig. 2 clearly shows that the surface morphology and structure of the divertor tile of DM2-2010 exposed to 6425 shots of plasma discharges (including 450 H-mode and 89 long pulse discharges) was quite different from the original one. Surface morphology of the original tile is a regular SiC crystal (Fig. 2a), while the exposed DM2-2010 tile shows a rough, dust-like structure (Fig. 2b). The surface compositions of the original tile and the exposed tile have been characterized using X-ray photoelectron spectroscopy (XPS), energy dispersive X-ray emission (EDX), and secondary ion mass-spectrometry (SIMS). The results demonstrated that the surface composition of the original tile is nearly the stoichiometric composition of SiC (52.74% C, 47.11% Si), but the deposited

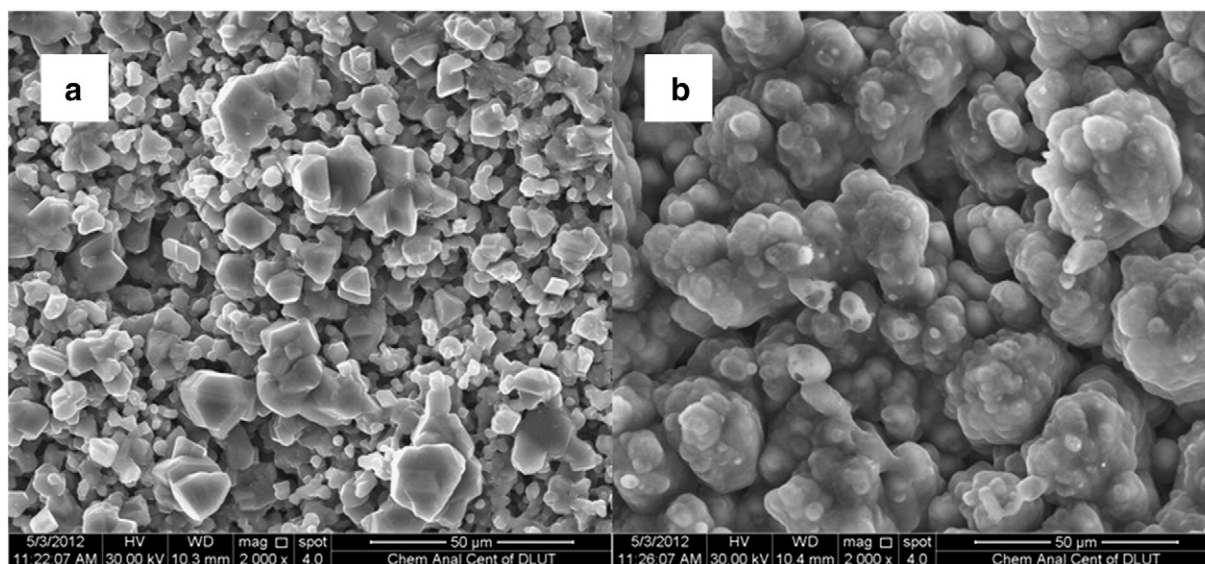


Fig. 2. (a) SEM picture of the original graphite tile; (b) surface topography of the DM2-2010 graphite tile after exposure.

layer of the exposed tile is composed mainly of Li, C, O and D due to Li being used as wall conditioning material and deuterium being used as the main plasma fuel.

3.2. Effect of pressure on the emission intensity of LIBS

The main challenge of the future LIBS diagnosis for tokamaks is the determination of the impurity deposition on PFCs in a vacuum environment. It is necessary to investigate the effect of pressure condition on the emission intensity. Fig. 3 shows the dependence of LIBS signals of H, C and Si for the original tile as a function of the pressure in the range of  $6 \times 10^{-6}$  mbar to 1000 mbar. The emission intensity initially increases with a decrease in pressure; a further decrease in pressure causes a decrease in the LIBS signal intensity. The maximum intensity achieved around at the pressure of a few mbar. This behavior can be explained by the competing processes of collisional excitation of species in the plasma and ablation mass from the tile. It is easily understood that the number of collisions per unit time decreases as the pressure decreases. On the other hand, the mass ablated increases at low pressure due to reduced plasma shielding [12]. In Fig. 4, the averaged ablation rate (AAR) shows a slight increase with decreasing pressure. The crater depth was measured using a surface profilometer (Kokasa ET4000M) with a machine resolution of 0.1 nm, and AARs at the different pressures were calculated.

3.3. Elemental depth profiling of divertor tiles

A systematic study of the elemental distribution of the divertor tiles has been carried out in our laboratory using LIBS. Fig. 5 shows the LIBS emission lines recorded after the first pulse using an energy density of 25 J/cm<sup>2</sup> at 5 mbar. As expected, the compositional differences between the original tile and the exposed tiles were noticeable. In the original tiles, the spectra present the composition of C, Si and Ti, with some contribution of hydrogen and oxygen. This is consistent with the composition of the doped graphite as SiC-coated tiles. In the exposed tiles, the superficial elements are dominated by high concentration of Li and low concentration of D and Fe. The deposition of Li came from wall conditioning, which played an important role in controlling the impurities and H recycling during EAST operation. Deuterium arose from fuel retention in the divertor tiles, and Fe was found to originate from the sputtering of the in-vessel component material (SS316). Moreover, the LIBS measurement revealed a trace amount of Mo impurity on the tiles exposed during the 2012 campaign. This may be due to the sputtering of the first wall, because Mo was chosen as the first wall material for the EAST.

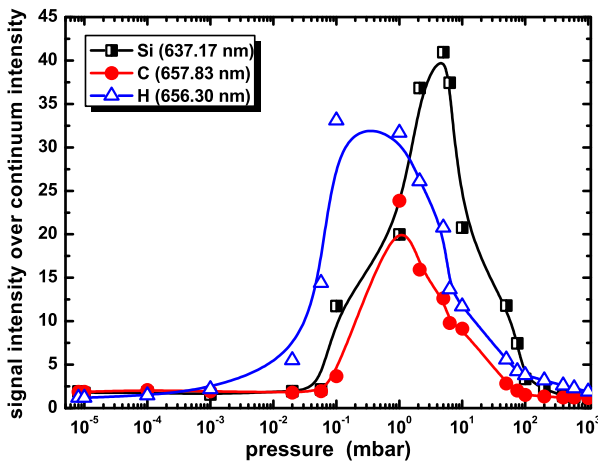


Fig. 3. Variation of the LIBS signal ratio to the continuum emission (bremsstrahlung) for Si, C and H as a function of the pressure. All spectra were taken at a fluence of 25 J/cm<sup>2</sup>.

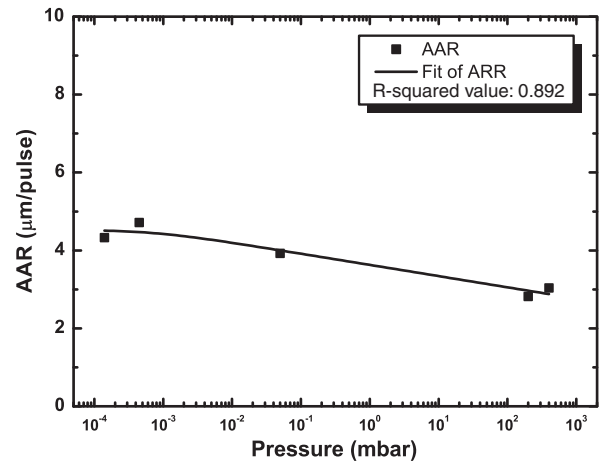


Fig. 4. Variation of AAR with pressure.

LIBS can convincingly provide a depth profile of the deposition layer compositions and the structural features of the divertor tiles using pulsed ablation. First of all, in order to study the retention of the deuterium in the exposed divertor tiles, the LIBS spectra of ID5-2012 with 1, 2 and 3 pulses were recorded as shown in Fig. 6. This shows that the signal of D declines sharply with increasing depth and indicates that the D is distributed only on the top layer of the exposed divertor tile. To measure the thickness of lithium deposition layer, 100 laser shots were fired on the same spot at a pressure of 5 mbar. This was sufficient to ablate the deposition layer from the

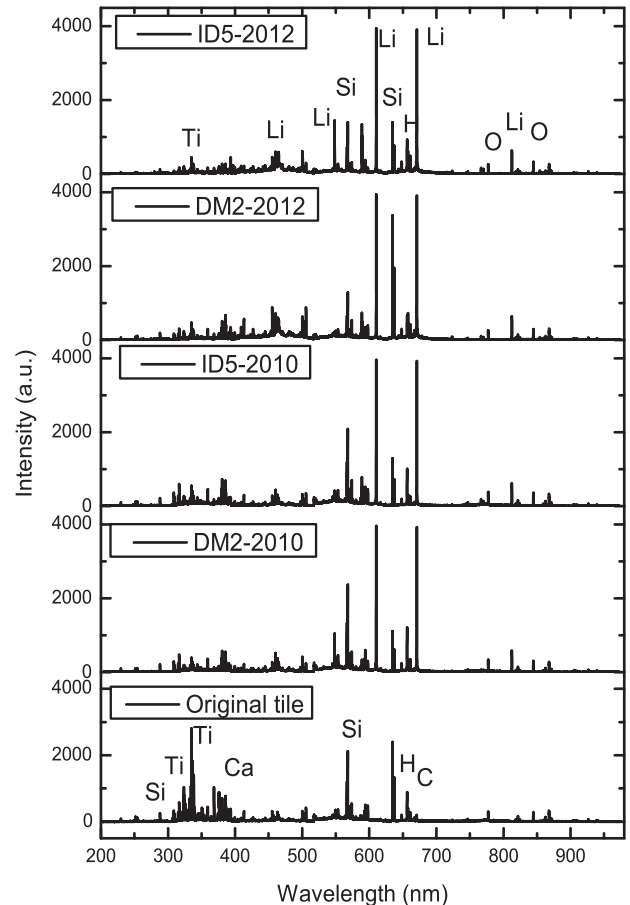


Fig. 5. LIBS spectra of ID5-2012, DM2-2012, ID5-2010, ID5-2010 and the original tile (top to bottom).

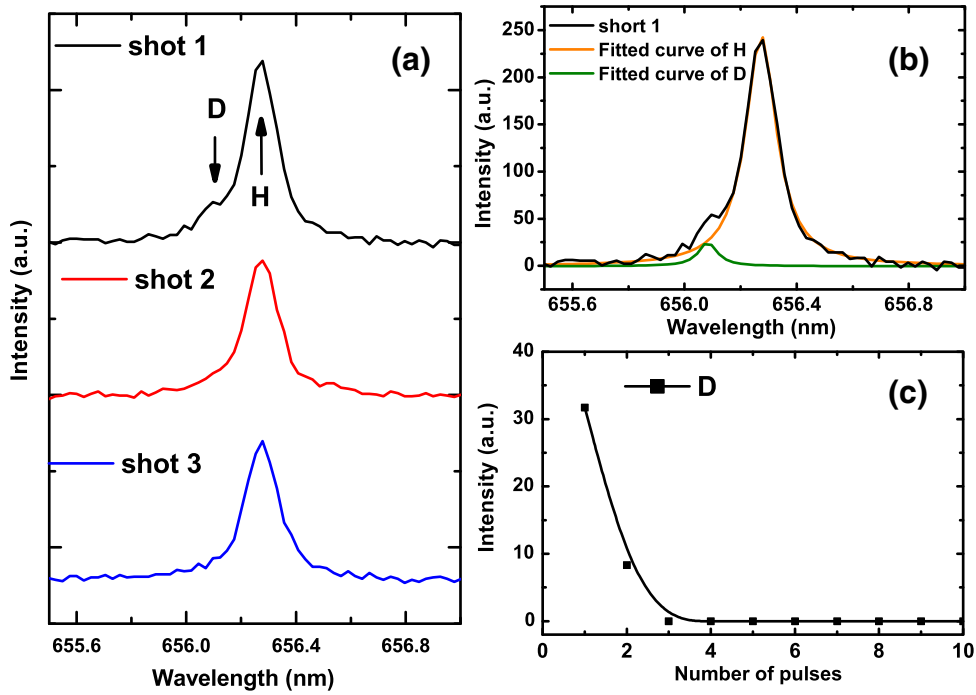


Fig. 6. (a) Deuterium and hydrogen spectra collected at pulses 1, 2 and 3 in the same spot during ablation of sample ID5-2012. (b) Enlarged view of spectrum collected at pulse 1 with fitted curves of H and D. (c) Evolution of the spectral line intensity of D.

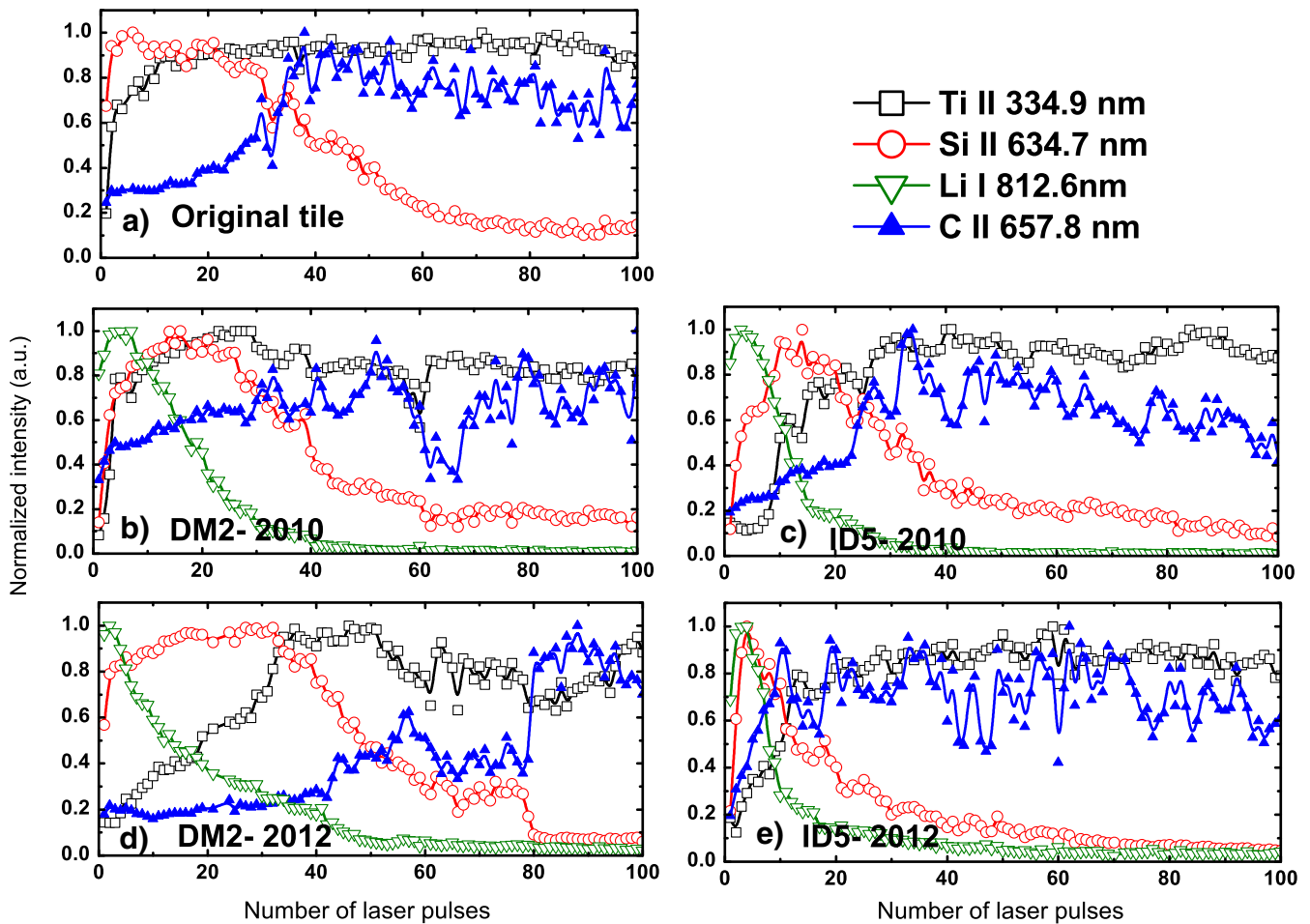


Fig. 7. Evolution of the spectral line intensities of ionic transition of Ti, Si, C and Li during ablation of the original tile and four exposed divertor tiles.

graphite substrate. The emission line of Li (I) at 812.6 nm was used for determining the LIBS depth profiles of the impurity in the deposited layer, and lines of Si II (634.7 nm) and Ti II (334.9 nm) were chosen to analyze the SiC coating and the substrate, respectively.

Fig. 7 gives the evolution of LIBS intensity as a function of the number of laser pulses. The profiles were normalized, which is a common procedure in depth profiling by LIBS [13]. As shown, the Li signal decreases with the number of laser shots, but the Si signal initially increases and then decreases with the ablation shots. Moreover, the signals of C and Ti increase and reach a plateau.

In Fig. 7a, it is remarkable that the Si signal becomes weak after 40 pulses for original tiles, but the Si signal does not disappear. This fact could be attributed to the non-uniform ablation process and the redeposition of the ablated materials. Because of the Gaussian-like energy distribution of the incident laser beam the divertor tile is ablated to produce a conical crater. The Si signal at the edge of the crater remains for a sequence of shots because the ablated region spreads over the edge gradually, leading to poor depth resolution at the interface [14]. In the future, we will use beam homogenizers to avoid non-uniform ablation. At lower pressure, a larger part of the ablated material could expand into the vacuum, thus resulting in negligible re-deposition.

An additional depth-resolved measurement was carried out at  $6 \times 10^{-6}$  mbar, and similar results (Fig. 8) were observed. Though the signals at  $6 \times 10^{-6}$  mbar are weaker, the line intensity evolution is in good agreement with the results obtained at 5 mbar.

In order to determine the thickness of the deposition layer accurately, a linear correlation analysis between the reference spectrum and the LIBS spectra from a depth sequence was carried out. The spectrum for the surface of DM2-2010 tile was selected as the reference spectrum and was recorded using a single laser pulse. It is associated with the deposition layer of the exposed tile. The spectrum obtained using a single laser pulse for the original tile surface was selected as the correlated spectrum and is associated with the substrate of the exposed tile. This spectrum was also recorded at the first laser shot ablating original tile.

The spectral regions (245–255, 286–291, 333–338, 600–680, 810–815 and 905–915 nm) were selected to record Si, Ti, Li, C and D, in the benefit of high signal-to-noise ratios and no interference from the other emission lines. The spectral regions of interest contained sufficient useful information about the “deposition layer” and the “substrate”. Finally the correlation coefficient was calculated by:

$$r = \frac{\sum_i (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_i (x_i - \bar{x})^2} \sqrt{\sum_i (y_i - \bar{y})^2}},$$

where  $x$  is the reference spectroscopic data set,  $y$  is a spectroscopic data set from the depth sequence,  $\bar{x}$  is the mean value of all  $x_i$ , and  $\bar{y}$  is the averaged value of all  $y_i$  [15]. The value of  $r$  lies between  $-1$  and  $+1$ , which can be used to indicate the correlation intensity between  $x$  and  $y$ . An  $r$ -value with near to 1 corresponds to complete positive correlation, when  $x$  and  $y$  spectra are from the materials with very similar chemical constitution. If the  $r$ -value is near zero, then the  $x$  and  $y$  are uncorrelated. Depth profiles are finally obtained by plotting the evolution of the linear correlation coefficient value for two reference spectra (of the deposition layer and the substrate) along the depth sequence of spectra, that is with the number of laser pulses or, if the averaged ablation rate (AAR) is known, the depth scale of laser ablation can be determined by the experiments [16]. The results of the correlation profiles are shown in Fig. 9. This figure revealed that the interface between the deposition layer and the substrate of the exposed divertor tile can be clearly identified.

In Fig. 9a, black curve revealed the correlated depth profiles associated to the reference spectrum of “deposition layer surface”. For ID5-2010 divertor tile, the high  $r$ -values in the depth from 0 to 9 pulses indicated that the thickness of the deposition layer could be about 40  $\mu\text{m}$  (measured by a surface profilometer). Meanwhile, the red curve revealed the correlated depth profiles related to the reference spectrum of “original tile corresponding substrate surface”. Values of  $r$  in the depth of 9 to 100 pulses were also near to 1, indicating the original tiles with the almost identical compositions. The vertical blue line determined the location of the interface between the deposition layer and substrate of ID5-2010 tile. In a similar manner, the vertical blue line in Fig. 9b showed the location of the interface between the deposition layer and substrate of DM2-2010 tile. The thickness of the deposition layer on DM2-2010 tile could be measured as  $\sim 50 \mu\text{m}$ . These results demonstrate that the  $r$ -values can easily distinguish the interface between thin impurity deposition layer and substrate of the divertor tile without the procedure of background correction and normalization.

## Conclusions

The main results concluded in this work, indicate that LIBS can be used in vacuum environments to monitor the deposition distribution in the deep layers of the plasma-facing materials of EAST. Monitoring the changes in the depth of the deposition layer can also provide useful information to evaluate if the PFCs need to be replaced. The influence of the LIBS signal intensity in pressure ranging from  $6 \times 10^{-6}$  to 1000 mbar was investigated. From the data we found that the signals were actually enhanced at a pressure of around a few mbar compared

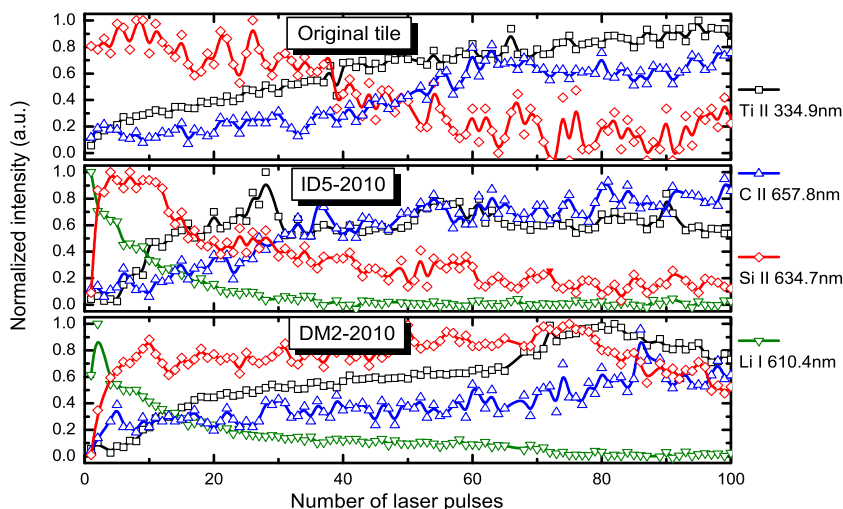


Fig. 8. Evolution of the LIBS intensities of ionic transitions of Ti, C, Si and Li for the original, ID5-2010 and DM2-2010 tiles at  $6 \times 10^{-6}$  mbar.

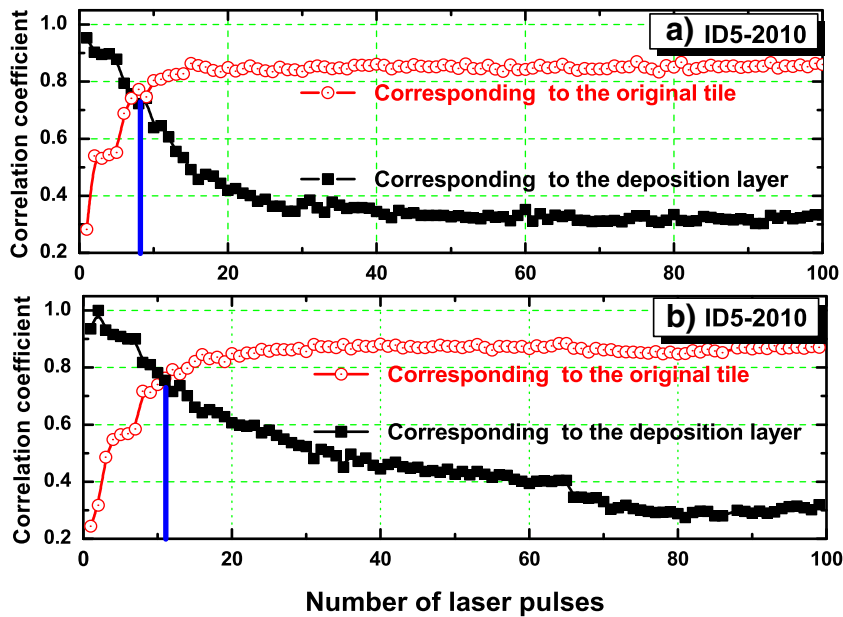


Fig. 9. Correlated depth profiles of the deposition layer surface and original tile surface. The vertical blue line indicates the location of the interface between the impurity deposition layer and the substrate.

to atmospheric and relatively low pressures. Though the signal intensity decreased significantly at lower pressure, the results of elemental depth profiling of divertor tiles at  $6 \times 10^{-6}$  mbar were in good agreement with the results obtained at 5 mbar. Linear correlation has been analyzed in our investigations to determine the thickness of multi-layers of divertor tiles at  $6 \times 10^{-6}$  mbar through the comparison of the LIBS spectra from the different depth sequences with the reference spectra. The results demonstrated the capability of this statistical method to distinguish the impurity deposition layer and the substrate of tiles without the need of background correction and normalization. The ability of linear correlation for improving the depth profile accuracy and identifying the interface localization is remarkable, but the previously ablated layers may still influence the elemental concentration measured by LIBS with the succeeding laser ablating shots due to Gaussian-like energy distribution of the laser beam, leading to poor depth resolution at the interface. A beam homogenizer is suggested to avoid the non-uniform ablation and to improve the depth resolution. Also additional studies should be performed to reduce laser fluence to achieve a better resolution in depth and to avoid ablating all deuterium retention in the first shot. This would be helpful for optimizing a LIBS system which can be used for in-situ monitoring of the deposition and erosion processes in the EAST.

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