Laser-induced breakdown spectroscopy to monitor ion cyclotron range of frequency wall cleaning Li/D co-deposition in EAST tokamak

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Abstract

In this paper, laser-induced breakdown spectroscopy (LIBS) **under magnetic field condition** has been **studied** in laboratory and EAST tokamak. The experimental results reveal that in helium ambient gas, the magnetic field significantly enhances the LIBS signal intensity (~ 3 times). The effect of time delay and laser fluence on the intensity of LIBS has been investigated for optimizing the signal to background ratio (S/B). The developed LIBS approach has been applied to monitor the cleaning performance of the first wall in the fusion device of EAST using the ion cyclotron range of frequency (ICRF). The experimental results demonstrate that the cleaning performance for Li/D co-deposition layer is effective under helium ambient gas. The removing rate of Li on the surface of W tile is faster than that on Mo tile in He-ICRF cleaning and the D/(D+H) ratio on Mo tile is higher by ~ 1.2 times than that on W tile. This work would indicate the feasibility of using LIBS to monitor the wall cleaning processes in EAST tokamak.

1. Introduction

Laser-induced breakdown spectroscopy (LIBS) has been regarded as a powerful optical emission spectroscopic technique for elemental analyses and characterization of the different types of substances included solid, liquid, aerosol and gaseous. In this approach, the sample material is vaporized and dissociated into ionic and atomic species which are excited to high energy levels. Spectra emitted by the excited species are applied to develop qualitatively and quantitatively analytical information. In the recent decades, LIBS has received increasing attention due to the advantages of low sample requirements, little sample preparation [1, 2], simple experimental arrangement and ability of fast multi-element and in-situ analyses [3, 4]. The potentialities of LIBS have been demonstrated in numerous fields of applications like cultural heritage [5], space exploration [6], and industrial control [7]. LIBS, as a fast, flexible in-situ monitoring technique can also be applied to the large volume vacuum device, such as the tokamak vessel [8-10]. Tokamak, such as ITER [11] and EAST [12], is a toroidal container that uses magnetic confinement to realize controlled thermonuclear fusion and solves the problem of human energy finally. However, during the tokamak operation, the heat flow and particles flux from core-plasma will interact with the plasma facing components (PFCs) and lead to a series of problems from impurity pollution on the first wall, such as impurities deposition, fuel retention and co-deposition etc., which will lead to the change of the surface morphology and composition of the first wall and affect the material properties and working life of the wall. Solving these problems is one of the most critical factors to realize the future ITER plan.

In tokamak devices, magnetic field is used to confine fuel plasma and controls it to the desired temperature. Therefore, study of the influence of magnetic fields on

LIBS diagnostic is quite insufficient. In this work, lithium alloy was chosen as sample material for investigating the influence of magnetic field on laser ablated Li-plasma. Many research groups have investigated the interaction of magnetic field with expanding plasma and observed some interesting phenomena, such as ion acceleration/ deceleration, spectral emission intensity enhancement, electron density and temperature increase, plasma plume splitting, instability and oscillating behavior and so on [13-16]. Ajai Kumar et al. [17-20] investigated the effect of magnetic field on the lithium plasma plume emission. Enhancement in the emission intensity and different structures of plasma plume in the presence of magnetic field have been observed. In our previous paper [21-23], the effect of magnetic field on laser-ablated plasma using the different sample (Al-Li alloy, tungsten, graphite) at the different ambient pressure have been investigated, and some interesting phenomena were also observed and discussed. Lithium is regarded as a potential first wall material of tokamak, due to its strong adsorption and inhibition capability to impurity gases, such as O₂, CO, N₂, and hydrogen isotopes gases. Moreover, Li can be used to inhibit the H impurities inside plasma, lower hydrogen recycling and improve the performance of plasma [24]. In EAST, lithium-wall conditioning associated with ion cyclotron range of frequency (ICRF) plasma cleaning was performed successfully, which resulted in the reduction of both residual impurity and H / [H+D] ratio as well as improvement of the performance of plasma [25]. Therefore, it is much desired to investigate the status of Li layer of the first wall of EAST device using LIBS technique. While, after a long time of EAST operation, lithium layer absorbed a large amount of impurities and hydrogen isotopes gases, which led to that the Li-layer lost the function of inhibition of impurities and reducing boundary particles recycle. So, the layer on the first wall must to be cleaned for EAST effective and safe operation.

Between discharges, ICRF wall cleaning can quickly remove the hydrogen isotopes retention on the surface of the first wall, due to the characteristic of being able to work under the environment of strong magnetic field. In EAST, as one of the main wall cleaning approaches, ICRF wall cleaning can effectively remove the

impurities deposited on the first wall **but** the cleaning performance depends on the working gases and deposited materials. Therefore, the study of cleaning processes of ICRF under the different working gases and **the** deposited materials is much desired.

LIBS, due to the advantages of multi-element analyses and in-situ monitoring, can be used to analyze wall material qualitatively and monitor the changes of the wall surface **condition** in real time [26-30]. In the present paper, firstly we **laboratorily** simulated the special constraints of fusion environments, including high vacuum, presence of the magnetic field and different ambient gases, to investigate the effects of magnetic field and the ambient gases on laser-produced plasma with the purpose of providing some useful information for **adopting** LIBS technique to monitor the ICRF cleaning processes of the first wall in EAST tokamak. Afterwards, we employed LIBS to EAST device for in-situ monitoring the ICRF cleaning performance of the first wall using helium ion cyclotron wall cleaning **approach**. Lithium deposition and D&H retention on the first wall (W and Mo tiles) have been measured in real time during ion cyclotron wall cleaning processes for the first time.

2. Experimental setup

The schematic diagram of the simulated experimental setup in the laboratory is depicted in Figure 1. In this experiment, an Nd-YAG laser (Brilliant Eazy, Quantel) with wavelength of 1064 nm was operated in the Q-switch mode at a 10 Hz repetition and 5 ns pulse-width with the maximum laser output energy of 200 mJ. The laser beam was focused onto the sample surface using a lens **with** focal length of 500 mm. The focus point of the lens was set a few millimeters behind the target surface. The mean diameter of the spot at the sample surface after 10 laser shots ablation was about 1.3 mm by measuring ablation craters. The pulse energy of 200 mJ corresponding to the fluence of 21.06 J/cm^2 was used.

An standard lithium-contained sample(Li 2.25%, Al 91.95%, , Mg 5.49%), in the form of strips of length of 20 mm, width of 5 mm and thickness of 2 mm, was fixed

on a sample holder. Two NdFeB permanent magnets in size of $50 \times 50 \times 25 \text{ mm}^3$ installed on both sides of the sample were used for creating a steady magnetic field. The spatial separation of the two magnets was set at 7 mm to generate a relative uniform field of 1.1 T measured by a Gauss Meter (LINK JOIN, LZ-643). During the plasma expanding, the size of plasma increased while the plasma plume was constrained spatially by the permanent magnets at the pressure of 2×10^{-5} mbar. Therefore, two cover glass plates (size: $22 \text{ mm} \times 22 \text{ mm} \times 0.17 \text{ mm}$) were stuck on the both sides of the sample holder to produce the same geometrically spatial constraint, which was used to ensure the same geometrical factor. The gap of two cover glass plates was also 7 mm. The sample was placed in the middle of the gap. The sample holder was mounted on a XY translation stage. The whole sample holder was placed in a stainless steel vacuum chamber which was equipped with quartz windows and was pumped to a minimum pressure of 1×10^{-5} mbar measured by a pressure gauge (Pfeiffer Vacuum, D-35614 Asslar, CMR 365).

The time-integrated LIBS signals (detection volume of 53.74 mm^3) were collected along the laser axis through a collection lens of focus length of 250 mm and was guided into an optical quartz fiber bundle coupled to a spectrometer (Ocean Optics Inc, US, spectral resolutions of 0.1 nm, and diffraction grating of 1200 grooves/mm). The detection system was equipped with seven linear silicon CCD array detectors, which provided a full spectrum in the range of 200-980 nm for each acquisition. The integration time (gate width) for collecting spectrum was set to 1 ms. Two-dimensional images of plasma plume were recorded by an intensified CCD (ICCD) camera (PI-MAX4, Princeton Instrument) interfaced with a computer and the minimum gate width of the ICCD camera was 3 ns. The data acquired simultaneously by all the seven channels were stored on another computer as shown in figure 1 for qualitative and quantitative analysis.

3. Results and discussion

3.1 The variation of emission intensity

By comparing the LIBS signals of Li plasma detected in the absence and presence of magnetic field, it has been observed that the spectral intensities are enhanced in several ambient gases (Argon, Helium and Air) with magnetic field at the pressure of 2×10^{-5} mbar shown in figure 2. But the factor of enhancement is different. The largest enhanced factor of Li atomic spectral intensity is observed by 3 times in He ambient gas. Pramod et al. [31] also observed the enhancement in intensity in the presence of magnetic field and ascribed it to the higher thermal conductivity of helium.

The laser-produced plasma consists of free electrons, ions and neutral particles. Under the external magnetic field condition, all kinds of charged particles are confined by magnetic field due to Lorentz force. This leads to the enhancement in emission intensity, due to laser plasma plume confinement. The evaporated atoms from the target material form a plume in the different background gases, i.e., Ar, He and Air. The effects of background gases on the plume cooling and expansion dynamics are different. It is seen from figure 2, the influence of magnetic field confinement on LIBS signal is conspicuous in He ambient gas. While without the magnetic field the strongest emission intensity is observed in Ar gas. At ambient pressure of 2.0×10^{-5} mbar, the pressure of laser-ablated plasma plume is calculated as ~ 100 mbar, much higher than the pressure of ambient gas. The nature of the ambient can't be ignored. Therefore, the lower ionization potential and thermal conductivity of Ar are thought to be the key factors to make the emission intensity enhancements in Ar, rather than in He and Air, Farid et al. [32, 33] also reported the similar results.

According to the theoretical work by Rai et al [34-36], the ratio (I_2/I_1) of plasma emission intensity in the presence and in the absence of a magnetic field can be written as:

$$\frac{I_2}{I_1} = \left(1 - \frac{1}{\beta}\right)^{-3/2} \left(\frac{t_1}{t_2}\right)^3 \quad (1)$$

where I_1 , I_2 is plasma emission intensity in $B=0$ and $B \neq 0$, respectively; $\beta = 8\pi nkT_e/B^2$, is the ratio of the kinetic energy of the plasma to the magnetic energy; t_1 , t_2 is the duration of spectral emission without and with the magnetic field, respectively; n , T_e and k are the amount of substance, electron temperature and Boltzmann's constant, respectively. Equation (1) indicates that enhancement ($\frac{I_2}{I_1} > 1$) in emission intensity is possible only when plasma β is low, assuming the emission durations equal ($t_1 = t_2$) in the $B=0$ and $B \neq 0$ conditions. So, the increment is significant if the plasma pressure is low enough. Without the magnetic field ($B=0$), at a pressure of 2.0×10^{-4} mbar, for the Ar ambient, the plasma temperature and electron density were much higher than in He [33]. With the magnetic field confinement ($B \neq 0$), the plasma temperature and electron density are increased [13-15, 21-22], and this is confirmed by the observed results. The pressure of plasma ($P = n_e k T_e$) is estimated to be 1.245×10^2 mbar in He gas and 1.497×10^2 mbar at Ar atmosphere at the laser power density of 21.06 J/cm^2 . Therefore, in He ambient gas, the plasma pressure was lower and the enhanced factor in emission intensity was the largest. This is due to its higher ionization potential (24.6 eV).

3.2 The effect of detection delay time and laser fluence on LIBS signal intensity

According to the section of 3.1, it can be acquired that the effect of magnetic field on LIBS signal is much stronger in helium ambient gas. In order to further understand the influence of ambient gases on experimental parameters, the effect of detection delay time and laser energy on LIBS' intensity will be investigated. Figure 3 shows the variation in the intensity of Li I 610.34 nm line from Li-plasma at three kinds of ambient gases, Ar, Air and He gas, with the different detection delay time in the absence and presence of magnetic field. An integration time of 1 ms was used to acquire the spectra and all the measurements were carried out at the pressure of 2.0×10^{-5} mbar. The laser fluence was set 21.06 J/cm^2 . As figure 3 shown, the emission intensities decline gradually with increasing gate delay time and the optimal detection delay was observed at 0 us. Therefore, the LIBS signal detection delay time was set to 0 us. The reason of emission intensity attenuating is the population decrease of the

excited atoms due to the reduction of temperature and electron density **when** the plasma plume expands and cools **down**. With the magnetic field, the **LIBS** intensity is enhanced. **Meanwhile**, the possibility of recombination radiation ($\propto 1/T^{3/2}$) rises. The externally magnetic field confines the plasma and leads to the increase of electron density. Therefore, the enhancement in **LIBS intensity** is probably due to the **increase** of the radiative recombination as a result of **raised** effective **electron** density due to magnetic confinement [37]. Figure 4 shows the variation in the emission intensities of Li I 610.34 nm line as a function of laser **fluence** at the different ambient gases (Ar, Air, He) with and without the magnetic field. At all the investigated background gases, the optical emission intensities increase monotonously with **raising** laser fluence both in the **B=0 and B $\neq$ 0 conditions**. This is due to the increase of the plasma density because of **enlarging** ablated mass and inverse bremsstrahlung as well as photoionization effects. Therefore, the results strongly indicate that the stronger laser power density leads to higher spectral intensity. It is obvious that the magnetic field can much significantly improve the sensitivity of LIBS detection system in helium background gas. While a high sensitivity of LIBS detection system is desirable for much **accurately** measuring the **status** of the first wall in tokamak device.

3.3 Monitoring H/D and Li deposition on the wall during ICRF cleaning processes in EAST

ICRF cleaning can effectively remove impurities **in** co-deposition layer on the first wall in EAST. However, so far, there is little **understanding** about ICRF wall cleaning mechanism under helium ambient gas. In this section, we firstly investigate the ICRF cleaning performance using helium as working gas and the cleaning processes on molybdenum and tungsten tiles.

In this study we applied LIBS to monitor the cleaning performance of the first wall in EAST using ion cyclotron wall cleaning under helium ambient gas with guided by our experimental research at the laboratory. A YAG **laser** ($\lambda = 1064 \text{ nm}$)

with the repetition of 10 Hz, pulse width of 5 ns, the laser beam fluence of 21.06 J/cm² has been used to LIBS measurements. The gate width was set 1 ms and the gate delay time was set at 0 us. Magnetic field in the EAST was 2.5 T and the pressure was 10⁻⁴ mbar. The first wall material in EAST is molybdenum alloy (TZM – Mo – Ti – Zr – C alloy). In order to study the impurities deposition and cleaning processes on the different wall materials, tagged tungsten tiles were installed on the groove of the first wall, and the size of tungsten tiles was 10 mm×10 mm×10 mm. Therefore, the target materials were molybdenum and tungsten tiles in our experiments. The mean size of the ablated craters after 20 laser shots ablation in LIBS experiments was about 1.1 mm in diameter. Fig.5 (a) and (b) show that the depth of Li deposition layer decrease with an increase of ICRF cleaning time and this implies that the impurities removal amount is proportional to the ICRF processing time. The results indicate that the ICRF cleaning performance is effective. After 180 minutes cleaning, the thickness of Li deposition layers almost no change with the further increase of ICRF cleaning time. The probable reason is that the mass of re-deposition of the impurities is equal to the amount of clearance. The amount of Li-retention on Mo tile is more than that on W tile and Li scavenging rate on the deposition layer of W tile is faster than that on Mo tile in He-ICRF cleaning. Figure 5 (c) presents the variation of D/ (D+H) ratio on Mo tile and W tile as a function of the ICRF cleaning time. This indicates that the D/ (D+H) ratio on Mo tile is higher by 1.15 ± 0.067 times than that on W tile. This means that Li wall-conditioning can absorb more D than H and the different wall substrate materials can influence the H/D absorption capacity. All these results demonstrate that LIBS is a useful tool to monitor the performance of wall cleaning in EAST tokamak.

4. Conclusions

A systematic study of the influence of the ambient gases at the pressure of 2.0×10^{-5} mbar on LIBS measurement in the magnetic field conditions has been carried out in laboratory as well as in EAST tokamak. The experimental results reveal that the ambient gas and magnetic field have a profound effect on the LIBS intensity. In the He ambient gas, the magnetic field enhances the LIBS intensity (~3 times). The

results were applied to measure the cleaning performance of the first wall in EAST by ion cyclotron wall cleaning under helium background gas. The ratio of D / (D+H) and lithium co-deposition on Mo and W tiles has been measured. The experimental results indicate that the cleaning performance is effective. Removing rate of Li on W tile is higher than that on Mo tile in He-ICRF cleaning. Meanwhile, D/ (D+H) ratio on Mo tile is higher by 1.15 ± 0.067 times than that on W tile. The value of D/ (D+H) is more than 0.72 ± 0.15 for both W and Mo tiles. This indicates that Li-wall conditioning absorbs more D than H. This work would illustrate the feasibility of adopting LIBS to monitor the wall cleaning processes in tokamak.

Acknowledgments

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Figure captions

Fig.1. Schematic diagram of the simulated LIBS' experimental setup at laboratory.

Fig.2. LIBS signal of Li I line at 2.0×10^{-5} mbar of (a) Ar, (b) Air and (c) He with and without a magnetic field. The data was taken in the same conditions and the measured spectra were averaged by 10 **single**-shot signals. The laser fluence was 21.06 J/cm^2 .

Fig.3. The intensity of the Li spectral line (610.34 nm) as a function of detection delay time at the different ambient gases (Ar, He, Air). The Li plasma was produced by nanosecond laser pulses with the **laser power density** of 21.06 J/cm^2 . All the data was averaged 10 laser shots.

Fig.4. The intensities of Li I 610.34 nm line with laser fluence at the different ambient gases with and without the magnetic field. The pressure was $2.0 \times 10^{-5} \text{ mbar}$ and delay time was set at 0 us. All the data was averaged 20 laser shots.

Fig.5. The depth of the Li-retention on the Mo tile (a) and W tile (b) at the different ICRF cleaning time as well as D/ (D+H) ratio on the first wall as a function of ICRF cleaning time (c).

Figures

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Fig.3.

Fig.4.

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