WHY IS LIBS THE FUTURE ONLINE ANALYTICAL TECHNIQUE FOR PLASMA-FACING MATERIALS OF THERMONUCLEAR REACTORS?

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1. INTRODUCTION

Currently, a global tendency of reducing carbon footprint is pursued by leading institutions and governments. Unlike other alternatives to fossil fuel burning, such as wind and solar sources, nuclear reactors provide a stable high energy output that can match the ever-rising demand. Even though nuclear fission plants have been well established (a total of 450 power plants in 31 counties), much of the research is being focused to nuclear fusion instead. An advantage over a fission reactor includes heavily reduced operational radioactivity and drastic reduction of radioactive waste. In fusion reactors deuterium and tritium are used as fuels. Deuterium is available in the seawater and tritium can be generated from lithium. For initiating the fusion reaction, a stable and very high temperature plasma is required. These conditions can be only achieved in magnetic confinement devices e.g. torus shaped tokamaks. One of such projects is The International Thermonuclear Experimental Reactor (ITER) that aims to generate fusion energy on a commercial scale in the range of 500 MW.

This research is focused on elemental analysis of plasma-facing components (PFCs) of the aforementioned ITER by means of laser-induced breakdown spectroscopy (LIBS). PFCs of a fusion device face harsh operating environment of high heat, radiation and neutron flux that lead to irreversible mechanical changes of the PFCs surface like cracking, blistering and erosion [Li 2016, Maury 2020, Merca 2011, Hai 2014]. During the high temperature plasma operation in tokamaks (despite of magnetic field confinement), the plasma interaction with walls - especially in the divertor and limiter zones - leads to the erosion of the PFCs. The eroded material migrates away and forms new mixed materials on the surface of the first wall or deposits in the form of flakes and dust in the cooler parts of tokamak.

Certain re-deposited materials exhibit higher fuel retention than original wall material and that is a serious problem for safety in the case of T fuel. In the case of ITER-like wall (tungsten divertor and beryllium principal wall), the re-deposition of Be/W layers together with O impurities leads to the more efficient trapping of D/T fuel than in the case of pure Be/W layers. In ITER, The maximum amount of permitted fuel within

the ITER reactor is up to 1 kg, so the retained T fuel in the first wall must be observed regularly to make sure its reliable operation with a complete fuel cycle [Oelma 2018].

Also due to the re-deposition of the impurities, the visibility of the mirrors and optical windows (essential components for the plasma diagnosis, imaging, and spectroscopy) is decreasing and their spectral transmittance and sensitivity is also changing. Their maintenance and protection is a serious issue for the operation of fusion devices [Mukhi 2009].

The study of material erosion and migration, the formation, release, and re-deposition of the dust are important issues. So, for the reliable and safe operation of the future fusion reactor, it is necessary to measure elemental compositions of all key PFCs periodically, for monitoring of impurities, its deposition pattern, and retained fuel.

The main advantages of laser-induced breakdown spectroscopy (LIBS) are in-situ and standalone analysis, no pre-treatment of the samples. LIBS analysis could be performed under vacuum, low-pressure, or up to atmospheric pressure environments and moreover magnetic field doesn't affect its performance. LIBS can be eventually combined with laser ablation (LA) based technique for the necessary cleaning process of the fusion device first wall [Fanto 2013, Schwe 2009].

2. RESULTS AND DISCUSSION

LIBS has been applied for the depth profile analysis of ITER-relevant materials, time-resolved and space-resolved analysis of PFCs, heating diagnostics of different PFCs, the study of multi-layered and calibrated samples, monitoring the features of the impurity layers deposited on the PFMs, in-situ and real-time diagnostics of the PFCs, etc [Brezi 2017, Maury 2017]. LIBS is an emission spectroscopic technique with the merits e.g., its speed, suitable for almost all kind of the samples, simultaneous multi-elemental analysis, minimal destructive nature (~1 μg material is required), analysis without sample preparation, the ability of contactless or remote analysis/sensing, immune to harsh environments, does not provide any interference itself, etc. Owing to its applicability and versatility, A robotic arm-based LIBS is being considered to monitor different parts of the reactor walls.

2.1. LIBS experimental set-up

Based on requirements in different fields, various LIBS set-ups such as conventional LIBS, back-collection LIBS, double pulse LIBS, etc. is being used for the elemental analysis and depth profile analysis of the samples [Maury 2020].

2.2. Calibration-free (CF) approach

For the CF-LIBS analysis [Ciucc 1999], more spectral lines for neutral and first ionization degree of the same element are necessary, if possible, emitted from different upper

levels. Otherwise, the evaluation of the electron temperature from the Boltzmann plot is doubtful. The transition probabilities for these lines should have good precision, and the spectral peaks should not be too weak, in addition, not to be in interference with other possible lines and not affected by self-absorption. We have performed a quantitative and depth profile analysis of the Be-W-D-based simulated layers (thickness 1.5–2 μm) on the Mo substrates for the H/D content. D content of the samples has been calculated using the CF-LIBS approach ~4.7% ± 2.9%, which is in good agreement with the other spectroscopic techniques for the same samples. We have also characterized Li-based coatings deposited on Ni-Cr based screws obtained after liquid metal experiments from the COMPASS vacuum chamber. After evaluation of plasma parameters using the multi elemental Boltzmann plot (MEBP) for different elements, impurities (e.g. Li, C, B) were quantified and depth profiling of Li content has also been verified.

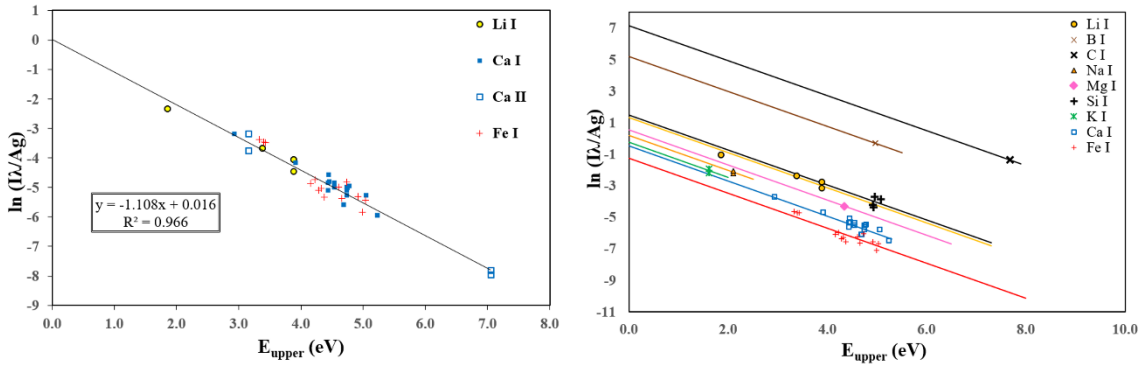

Figure 1. MEBP for the elements Li I, Ca I-II, and Fe I, and Boltzmann plot for Li I, B I, C I, Na I, Mg I, Si I, K I, Ca I, and Fe I spectral lines using the evaluated Te from the MEBP (for one of the screws from COMPASS Li campaign) [Veis 2020a].

CF-LIBS approach is also important for the control of impurities and dust pollution in tokamaks or linear fusion devices. Finally, this LIBS based monitoring is important for the knowledge of the pollution level caused by previous specific campaigns (e.g. previous carbon-based first wall, special limiter, or test with liquid metals, etc.). More precise T_e evaluation from W SB plots by enlarging LIBS analysis down to VUV range was recently proposed [Pribu 2016].

2.3. Molecular bands and LAMIS

Emission of molecular bands of diatomic radicals appears significantly later in the decay of laser induce plasmas, at the moment, when atomic lines start to disappear. So, the molecular emission measurements should have to be done separately from the atomic emission measurements. The perspective of the molecular emission spectroscopy in the fusion reactors wall diagnostics is the ability of an efficient isotopic analysis, due to the fact that the molecular lines isotopic shift is several order of magnitude higher than the atomic line isotopic shift. This idea of so-called Laser Ablation Molecular Isotopic

Spectrometry (LAMIS) was proposed in 2011 by Russo et al. [Russo 2011] and a review paper about the LAMIS method was published in [Bolsh 2016].

A possible application in LIBS fusion research is for the efficient quantification of relevant isotopes (e.g. D/H [Sarka 2013, Bolsh 2017], B [Mao 2011], C [Dong 2013], N, etc.) in laser-induced plasma after the conversion to molecular radicals as OH/ OD, BO, C_2, CN and others. For separation of hydrogen isotopes H/D (and in the future also T), the proposed molecular OH/OD (A-X, Δv=+1) transitions at 281 nm and 287 nm respectively have large separation (6 nm) of both isotopes bands [Russo 2011].

2.4. Depth profile analysis

LIBS has been found promising for the depth profile analysis by recording a successive number of LIBS spectra at the same position of the samples (especially of the analysis of thin layers and deposited impurities). Variation of the spectral signal with the number of laser shots represents depth profiling of the sample and provides information about thickness of the sample layer and ablation rate. LIBS depth profile analysis has been applied for different materials from fusion devices (JET, WEST, COMPASS, W7-X etc.). Veis et al. [Veis 2020b, Sucho 2017a] have applied depth profile analysis and quantification of the elements in Be/W(D) and Dwivedi et al. [Dwivedi 2020] have applied the same method for Be-O-C-D mixture samples and found in good agreement with other established technique like SIMS, NRA and TDS for D retention. Depth profile analysis of LiSn alloy by CF-LIBS was proposed in [Sucho 2017b].

Quantitative LIBS elemental and depth profile analysis of prepared mixed fusion relevant layers or post-mortem analysis of samples exposed in linear machines (Magnum-PSI, PSI-2) or tokamaks (JET, WEST, ASDEX, Compass….) were or could be used for validation by several off-line techniques like AAS, ICP-MS, spark discharge AES, NAA, RBS, NRA, TDS, SIMS, XRF and others. Most of the techniques are time-consuming and need special sample preparation, which is a drawback for in-situ, and online experiments.

2.5. Fuel retention and separation of the hydrogen isotope lines

For safety and control purposes of fusion devices, in-situ analysis of fuel retention is very important. In ITER, A remote-LIBS has been considered the most promising technique for the detection of fuel retention in plasma-facing walls and divertor materials regularly. Paris et al. [22, 23] have studied erosion, deposition, and fuel retention in fusion-related materials using LIBS and compared with other postmortem analysis techniques like NRA, RBS, and SIMS.

At high pressure, in laser-induced plasma, H_α (656.3 nm) and D_α (656.1 nm) lines are partly overlapped due to the Stark broadening effect. Large gate delay can reduce this overlapping, but spectral lines are with a low S/N ratio. Suitable conditions for resolving these lines has been optimized. At atmospheric pressure, suitable gate delay was optimized >2 μs, while vacuum conditions provide well-resolved D_α and H_α lines.

3. CONCLUSION

Owing to its inherent advantages (in-situ and standalone analysis, no pre-treatment of the samples, possible combination of laser diagnostics and laser cleaning process), LIBS, LIBS-based techniques are useful to diagnose and/or quantify the fuel retention, the impurities and the depth profiling of coatings and redeposited mixed materials related to the fusion devices.

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