On the use of laser-induced breakdown spectroscopy to determine the fractional abundances of carbon ions in the laser plasma plume

M. Naiim Habib[1], Y. Marandet[2], L. Mercadier[3], Ph. Delaporte[3], C. Hernandez[1], N. Gierse[4], M. Zlobinski[4], P. Monier-Garbet[1], C. Grisolia[1], B. Schweer[4], A. Huber[4], V. Philipps[4]

[1]*CEA, IRFM, F-13108 Saint-Paul-lez-Durance, France.*
[2]*PIIM, CNRS-Université de Provence, Marseille, France.*
[3]*Laboratoire Lasers, Plasmas et Procédés Photoniques, Marseille, France.*
[4]*IEK-4, Association EURATOM-FZJ, TEC, Jülich, Germany.*

1. Introduction

In tokamaks, spectroscopic measurements are routinely used to monitor particle fluxes into the plasma. To check if these measurements can be used for erosion determination, a method which consists in injecting in the line of sight of the diagnostic a known particle source created by laser ablation, during plasma operation, has been proposed. A feasibility study of this technique is presented in [1], where preparative laboratory experiments were realised to characterise a laser produced carbon particle source. However, in order to infer the particle source in magnetised plasmas, the relative quantities of atoms and ions have to be determined. In this contribution, we investigate the possibility to determine this ratio by analysing the spectra emitted by the laser-induced plasma. First results on the analysis of spectroscopic emission in the TEXTOR tokamak, where carbon particles were injected during plasma operation using the laser ablation system [2], are then presented.

2. Laboratory experimental setup

The experimental set-up is described in [1]. A polycrystalline graphite target was irradiated at normal incidence by an Nd:YAG laser (1064 nm, 5 ns, 10 Hz), with a maximum fluence of about 50 J/cm², under an argon pressure of 0.5 mbar (pressure relevant to tokamak edge plasmas ~ 0.4 mbar). Laser-Induced Breakdown Spectroscopy (LIBS) measurements, which consist in analysing the optical emission of the laser-produced plasma, were performed. Spatially integrated spectra were acquired for different delays after the laser pulse, with an acquisition gate of 20 ns duration. The plasma plume was imaged along an axis at an angle of 15° to the surface normal onto the entrance of an optical fiber linked to an Echelle spectrometer, equipped with an intensified camera. The signal was accumulated over several hundreds of laser shots, in order to increase the signal-to-noise ratio. C, C^+ and C^{2+} lines, as well as C_2 Swan molecular band and H_α line at 656.28nm were identified on the experimental spectra. The presence of this latter line is due to hydrogen absorbed by the carbon sample in ambient air.

3. Modelling of the experimental spectra

To be able to estimate the amount of particles that will effectively penetrate into the tokamak plasma edge, the relative quantities of atoms and ions have to be determined. Indeed, charged particles are expected to undergo prompt redeposition, because they will follow the magnetic field lines, which are almost tangent to the limiter surface.

The determination of the ionisation degree of a laser-induced plasma from the spectra measured by LIBS is often based on the assumption of local thermodynamic equilibrium (LTE) [3]. According to the McWhirter criterion, stationary and homogeneous plasmas in LTE satisfy the following relation: n_e (cm^{-3}) $\geq 1.6\times10^{12}\ T^{1/2}(\Delta E)^3$, where ΔE (eV) is the largest electronic transition considered (for carbon, $\Delta E \approx 6.5$ eV), and T (K) is the electron temperature. The electron density n_e, determined from the Stark broadening of H_α Balmer line using the PPP code [4], is 2.6×10^{15} cm^{-3} for a delay of 200 ns after the laser pulse. At this delay time, the apparent excitation temperature of CII species determined from the Boltzmann diagram is $T_{exc} \approx 24900$ K (correlation to the linear fitting $R^2 = 0.66$), while the apparent ionisation temperature deduced from the Saha-Boltzmann plot is $T_{ion} \approx 22500$ K ($R^2 = 0.95$). As a result, in our case the minimum density required for LTE is about 7×10^{16} cm^{-3}, so that LTE should not be satisfied. Although T_{exc} is close to T_{ion}, the points of the Boltzmann diagram are not well aligned, which supports the non-LTE hypothesis.

The emissivity of the 9 carbon lines identified on the experimental spectra (see Table 1) are modelled using the following relation: $\varepsilon_\lambda^z = n_e\left(n_z PEC_{exc}^{z,\lambda}(n_e,T_e) + n_{z+1} PEC_{rec}^{z+1,\lambda}(n_e,T_e)\right)$. In this expression, λ is the wavelength of the considered transition in a z ionisation state, n_e and T_e are respectively the electron density and temperature, $PEC_{exc}^{z,\lambda}(n_e,T_e)$ is the excitation photon emissivity coefficient, $PEC_{rec}^{z+1,\lambda}(n_e,T_e)$ is the recombination photon emissivity coefficient, and n_z and n_{z+1} are respectively the population density of z and $z+1$ ionisation states. The PECs, which depend on n_e and T_e, are given in ADAS [5], and synthesise all the processes of population and depopulation of atomic levels. A genetic algorithm [6] is used to adjust the maximum intensity of the various spectral lines by least squares minimisation.

Species	λ (nm)	A_{ul} (s^{-1})	E_l (eV)	g_l	E_u (eV)	g_u
CI	247.856	2.80×10^7	2.684011	1	7.684766	3
CII	251.206	5.61×10^7	13.720780	4	18.654857	6
CII	274.649	4.36×10^7	16.331739	2	20.844686	4
CII	392.069	1.27×10^8	16.333122	4	19.494539	2
CII	426.726	2.38×10^8	18.045986	6	20.950643	8
CII	658.288	3.66×10^7	14.448826	2	16.331739	2
CII	723,642	4,18.10^7	16.333122	4	18.045986	6
CIII	229.687	1.38×10^8	12.690035	3	18.086334	5
CIII	465.025	7.25×10^7	29.534646	3	32.200085	3

Table 1: Wavelength (λ), radiative decay (A_{ul}), energy (E) and statistical weight (g) of upper (index u) and lower (index l) levels of the selected carbon lines, taken from the NIST database (http://www.nist.gov/pml/data/asd.cfm).

As the laser-induced plasma is non-homogeneous, we assume that the plume can be divided in two regions having different temperatures, corresponding respectively to the hot plasma edge and to the cooler plasma core. Indeed, at a low pressure, the most excited species are the fastest and are located in the plasma front in the first hundreds of nanoseconds [7]. These 2 temperatures and the fractional abundances of carbon ions are set as free input parameters of the genetic algorithm.

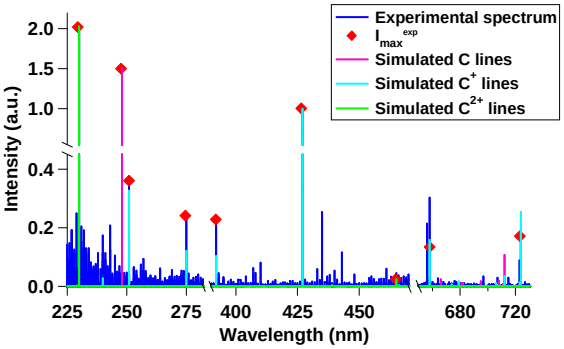

Fig.1: Fit of the experimental spectrum (measured 200 ns after the laser pulse).The red diamonds (I_{max}^{exp}) indicate the maximum intensity of the selected carbon lines on the experimental spectrum.

As shown on Fig.1, there is a good agreement between the experimental and the simulated spectra. In this case, the fractional abundances of C and C^{2+} are respectively 0.18 and 0.82, while the abundances of C^+ and C^{n+} with $n \geq 3$ are negligible. The temperatures obtained for this fit are of the order of 1.3 eV and 6 eV. In plasmas generated by nanosecond laser ablation, recombination processes prevail over ionisation processes [8]. C^+ lines observed on our experimental spectrum are thus probably mostly due to recombination of C^{2+} ions with electrons, the temperature being too low to observe significant emission from excitation of C^+ ions. These results are the subject of ongoing work.

4. First *in situ* experiments in the TEXTOR tokamak

The experimental setup described in [2] was used to realise in situ experiments in the TEXTOR tokamak. A polycrystalline graphite target, located at 4 cm outside the last closed flux surface, was irradiated by an Nd:YAG laser (1064 nm, 7 ns) at a fluence of 7.2 J/cm². Carbon particles ($N_{abl} = 2.9 \pm 0.4 \times 10^{18}$) were injected by one laser shot during a plasma discharge, with a central electron density $n_e = 1.5 \times 10^{19}$ m^{-3}, a toroidal magnetic field $B_t = 2.25$ T, and a plasma current $I_p = 350$ kA. A neutral beam injector was used to heat the plasma. An Echelle spectrometer in the visible range allowed monitoring the interaction between the laser-injected particles and the tokamak plasma. Fig.2 compares the spectra recorded (125 ms integration time) with and without injection of carbon particles. The emission intensity of CII, CIII and C_2 species increases in the presence of laser-injected carbon particles. With the knowledge of the amount of particles effectively penetrating into the plasma, for a given spectral line of wavelength λ, the ratio $\alpha(\lambda) = (I_{plasma+laser} - I_{plasma})/N_{abl}$ allows to determine the link between the measured intensity and the amount of eroded particles, and thus to calibrate

regularly *in situ* the spectroscopic measurements. As the laboratory experiments presented in section 3 were realised in ablation conditions different from those applied in TEXTOR, the fractional abundances of carbon ions cannot be deduced easily.

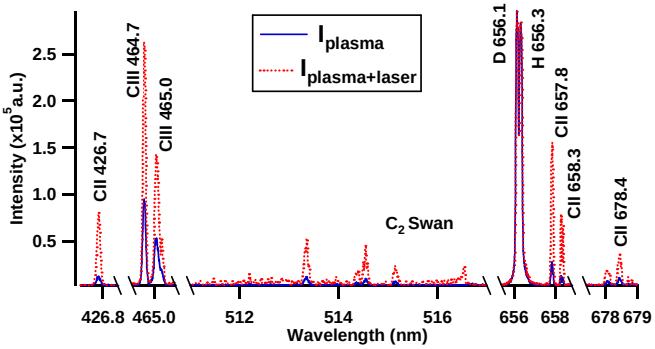

Fig.2: Comparison of the spectra obtained with the Echelle spectrometer in TEXTOR with the tokamak plasma alone (I_{plasma}) and in presence of carbon particles injected by laser ablation ($I_{plasma+laser}$).
$n_e = 1.5\times10^{19}$ m-^{3} ; $B_t = 2.25$ T ; $I_p = 350$ kA.

New laboratory experiments in the TEXTOR ablation conditions and/or *in situ* experiments without plasma but in presence of the magnetic field are therefore required to be able to interpret correctly the experimental data, and to estimate the amount of eroded material.

5. Conclusions and outlook

The model used to simulate the laboratory experimental spectra to evaluate the fractional abundances of carbon ions in the laser plasma plume gives encouraging results. However, further investigations on radiation transport along the line-of-sight and on the temperature spatio-temporal evolution are needed. The first in situ experiments realised in TEXTOR allowed to observe the plasma plume as well as the interaction between the injected particles and the tokamak plasma, and to understand qualitatively the influence of the laser-injected particles on the spectroscopic measurements. Further laboratory and *in situ* experiments are needed to better understand the influence of the various parameters and to obtain more quantitative results.

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