

Abstract : A spectroscopic study of the plasma plume created by a laser beam on the surface of a CaCl₂ aqueous solution is presented. Optical emission spectra are recorded and temporally analyzed. The electron number density is determined from the Stark broadening of the nitrogen NI 746.8 nm atomic line and the temperature profile is obtained from the relative intensity of OI lines. To take into account the possible occurrence of the self-absorption phenomenon, the escape factor of each atomic line is computed and used to correct the measured line intensities in order to obtain reliable temperature and density profiles.

Experimental Set-up

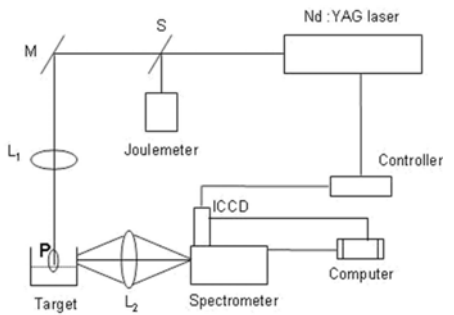


Figure 1: Experimental setup for laser ablation in aqueous solution. M: mirror high-reflectance coated at 532 nm, S: beam splitter, P: plasma plume, L1: converging lens, L2: imaging lens.

The plasma is formed by focusing with the lens L₁ a pulse Nd: YAG laser on the surface of a CaCl₂ (5x10⁻⁴ mol/litre) aqueous solution. The focalisation point is located 1 mm under the surface of the solution. The light emitted by the plasma is collected on the entrance slit of a 0.50m focal length spectrometer. The spectrum is recorded by a time gated ICCD camera (Istar Andor DH734-18F-03) equipped with a CCD matrix of 1024x1024 pixels (13x13 mm). The camera and the laser are controlled by a pulse generator (Systron Dooner). The spectra are recorded by the camera using an exposure time of 10 ns for each measurement.

Diagnostic methods

Selected spectral lines :
 ⇒ OI 777.3 nm and OI 715.7 nm for temperature
 ⇒ NI 746.8 nm for electron number density
 ⇒ Hα 656.3 nm

Temperature : relative intensity
 In LTE plasmas, the temperature could be determined with the relative intensity method. T is simply calculated from the ratio of two line intensities of the same chemical element (oxygen in the present study):

$$T = \frac{E_m - E_l}{k_B} \times \left[\ln \left(\frac{I_{ik} g_m A_{ul} \lambda_{ik}}{I_{lm} g_l A_{ul} \lambda_{lm}} \right) \right]^{-1}$$

Electron number density : Stark broadening
 The electron number density is usually deduced from the Stark broadening of an atomic line. Since several nitrogen lines are observable in the recorded spectra because of the pumping of the surrounding gas (air), the electron number density was finally determined from the Stark broadening of the nitrogen NI 746.8 nm line:

$$\delta\lambda = 2 \left(1 + 1.75 \times 10^{-4} n_e^{1/4} A \left(1 - 0.068 n_e^{1/2} T^{-1/2} \right) \right) w_e 10^{-16} n_e$$

The TLE assumption is unnecessary with this technique because it is independent of the pressure and of the plasma composition.

Self-absorption theory

With the assumption of local thermodynamic equilibrium, in the case of an optically thin, homogeneous, isothermal plasma, the total radiance (W m⁻² sr⁻¹) of a particular line is given by

$$I_{\lambda_0}^{thin}(T) = L_{\lambda_0}(T) \cdot R_p \cdot \int_0^{\infty} k_{\lambda}^{\prime}(T) d\lambda$$

where λ₀ is the central wavelength of the transition, L_{λ,0} is the Planck distribution function (W.m⁻².sr⁻¹.m⁻¹) for the blackbody radiation, R_p is the radius of the emitting plasma region and K_λ is the monochromatic effective absorption coefficient (m⁻¹) corrected for the effects of stimulated emission. If the self-absorption phenomenon is significant, the real line radiance can be written as

$$I_{\lambda_0}(T) = I_{\lambda_0}^{thin}(T) \cdot \Lambda_r(\lambda_0)$$

Where Λ_r is the so called 'escape factor'. This dimensionless parameter whose value lies between 0 and 1 is defined as the ratio between the real radiation flux escaping from the plasma and the radiation flux in the optically thin case. It is given by

$$\Lambda_r(\lambda_0) = \int_0^{\infty} P(\lambda) \exp[-\tau_0 \frac{P(\lambda)}{P(\lambda_0)}] d\lambda$$

where τ₀ is the monochromatic optical thickness: τ₀ = k_λ'(T) · R_p
 ⇒ The calculation of the escape factor requires the knowledge of the line profile resulting from the different broadening mechanisms: Doppler effect and pressure (Stark, resonance and Van der Waals broadening). in the most general case, Doppler and pressure broadening mechanisms are both significant and the line shape should be described by a Voigt profile.

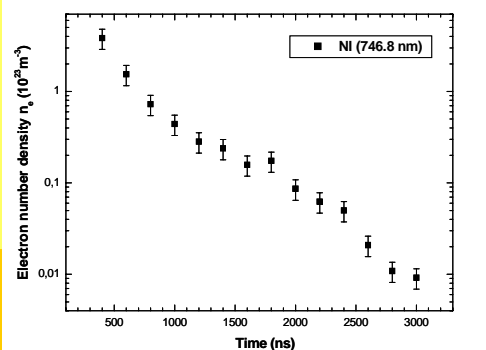


Figure 2: Electron number density derived from the Stark broadening of the NI 746.8 nm line

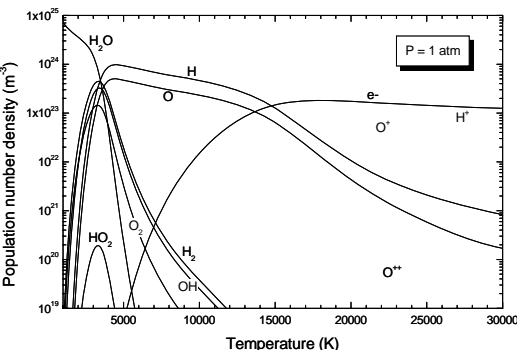


Figure 3: Water plasma equilibrium composition.

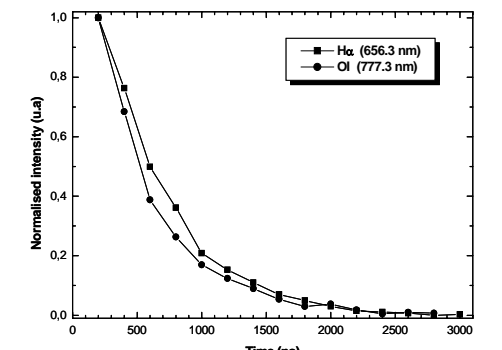


Figure 4: Experimental integrated line intensities as a function of time.

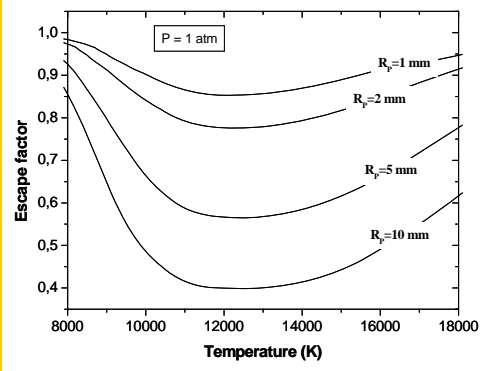


Figure 5: Escape factor of the OI 777.3 nm line for several plasma radius.

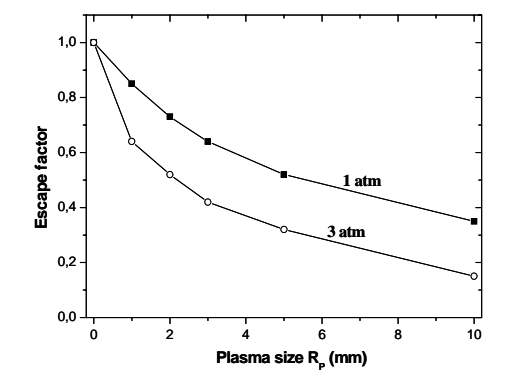


Figure 6: Escape factor of the OI 777.3 nm line as a function of pressure and plasma radius.

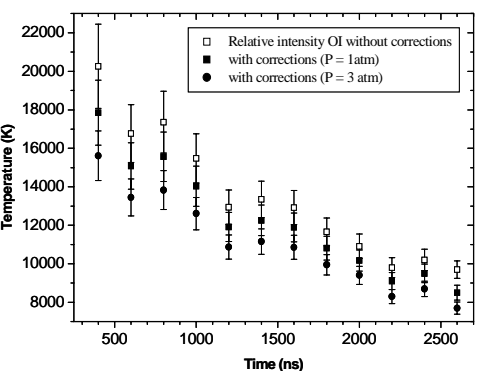


Figure 7: temperature profiles with and without self-absorption corrections.