LIF measurements of OH in a plasma-jet plume and oxidation of fatty acid methyl esters.

L.M. Martini[1], M. Scapinello[2], G. Bortolotti[1], G. Dilecce[1,3], M. Scotoni[1] and P. Tosi[1]

[1] *Department of Physics, University of Trento, via Sommarive 14, IT-38123, Trento, Italy*
[2] *CNR-IMCB, UOS Trento, via Sommarive 14, 3IT-8123, Trento, Italy*
[3] *Istituto di Metodologie Inorganiche e dei Plasmi-CNR, via Orabona 4, IT-70125, Bari, Italy*

Abstract: Reactive oxygen species are effectively generated in a radio-frequency plasma-jet, by using He/H_2O as feed gases. By laser induced fluorescence, we have measured the OH density in the plasma plume impinging on a liquid target of fatty acid methyl esters. This allows to investigate the role of OH in the oxidation of the target.

Keywords: laser-induced fluorescence, non-thermal plasma jet, reactive oxygen species

1. Introduction

Atmospheric-pressure plasma jets (PJs) allow to generate reactive oxygen species (ROS), such as OH, O and $O_2(a^1\Delta_g)$ radicals. For this reason, these discharges are of interest in sterilization and other biomedical applications [1, 2]. From the viewpoint of basic research, PJs can be utilized to investigate radical-initiated oxidation processes [3]. We used laser induced fluorescence (LIF) to measure the OH concentration in a radio-frequency discharge of He/H_2O. The jet plume impinges over the surface of a liquid target, composed of fatty acid methyl esters (FAMEs). OH radicals produced in the discharge may thus trigger the oxidation of unsaturated FAMEs.

2. LIF

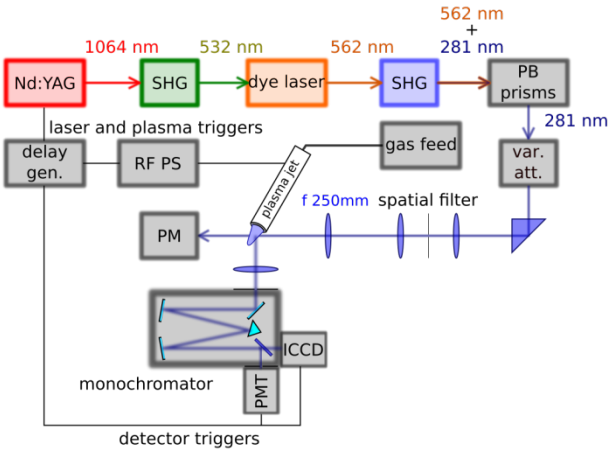

Fig. 1. Schematic representation of the LIF experimental setup. SHG, second harmonic generator; PB prisms, fundamental separator; var. att., variable optical attenuator; PM, thermopile-based energy detector

The LIF set-up is shown in Fig. 1. It is composed of a dye laser (TDL50, Quantel with Rhodamine 590 chloride) pumped by a Q-switched Nd:YAG laser (YG580, Quantel). The dye laser is frequency doubled using a KDP crystal harmonic generator. The UV-beam energy at 2810 Å, after fundamental separation, is around 0.53 mJ/pulse. The energy of the beam is controlled by an variable optical attenuator based on Fresnel reflection. The attenuated beam is spatial filtered prior to be focalized inside the target area by a f = 250 mm lens. The energy of the laser beam is recorded by a single-channel silicon-based thermopile (DX-0576, Dexter research center). The fluorescence signal is collected in the scatter arrangement by a f = 200 mm lens and injected in the detection system by a f = 75 mm lens. A Shamrock 303i 300 mm focal length monochromator, equipped with two gratings (1200 gr/mm blaze 300 nm and 600 gr/mm blaze 300 nm) is used to separate the spectral components of the fluorescence signal. The monochromator is coupled with two different detectors: a 1024x1024 px intensified CCD (ICCD) (DH334T-18U-03, Andor) and a getable photomultiplier tube (PMT) (8575, Burle Electron Tubes). The ICCD is used to record the spectrum of fluorescence signal, while the PMT serves to record the temporal evolution of the LIF signal. We adopted the classical excitation-detection scheme that involves transitions of the 3064Å system [4]:

$$OH(X^2\Pi, v, N) + hv_L \rightarrow OH(A^2\Sigma^+, v', N') \rightarrow OH(X^2\Pi, v'', N'') + hv_F \qquad (1)$$

with $\upsilon = 0$, $\upsilon' = \upsilon'' = 1$, and N is the rotation quantum number. Ground state rotational temperature was determined by measuring the LIF excitation spectrum in the small spectral region including the group of lines $Q_{12}(1)$ at 2829.21 Å, $Q_2(1)$ at 2829.23 Å, $Q_1(6)$ at 2829.27 Å, $Q_{12}(3)$ at 2829.31 Å and $Q_2(3)$ at 2829.37 Å. OH density was calculated using the $Q_2(3)$ line. The model used for interpreting LIF outcomes is described in [5].

3. Discharge design

The plasma source (see Fig. 2) consists of a tungsten needle of 2 mm diameter that acts as high voltage electrode, surrounded by a quartz tube (3 mm inner

diameter, 5 mm outer diameter) where a mixture of He and H_2O flows.

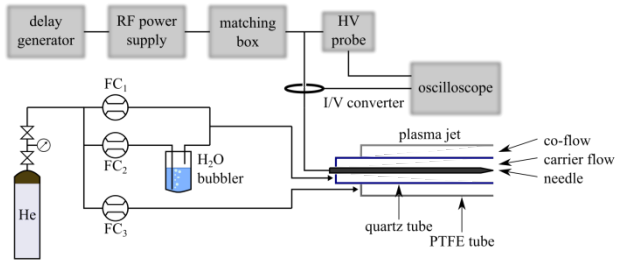

Fig. 2. Schematic representation of the radio frequency plasma jet. FC1,2,3 mass flow controllers

In order to prevent contamination of the plasma plume by the outside atmosphere a PTFE tube (6 mm inner diameter, 7 mm outer diameter) concentric to the needle is used to inject a co-flow of pure He. The He/H_2O mixture is obtained by injecting a fraction of the gas carrier inside a bubbler filled with distilled water at room temperature. The grounded electrode (an aluminum cylinder of 20 mm diameter) is placed 11(1) mm below the tungsten needle tip. The target is placed inside a small PTFE cylindrical vessel on the grounded electrode. A 500 W RF power supply (RF5S, RFPP) working at 13.56 MHz generates the discharge. Impedance matching between the power supply and the plasma jet is achieved by using an automatic matching network (AM5, RFPP).

4. Measurement of the electrical power

To measure the power dissipated in the plasma, we used a voltage probe (P6015A, Tektronix) and a high bandwidth (200 Hz to 500 MHz) I/V converter (CT-C1.0, Magnelab). Their signals were recorded and digitized by an oscilloscope (WaveSurfer 104MXs-A, 1 GHz, 5 GSps,

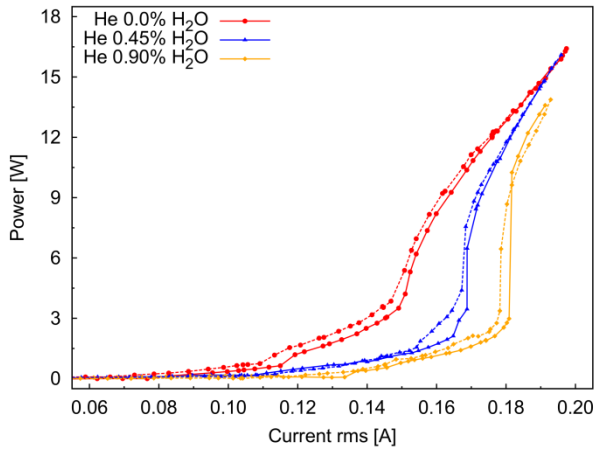

Fig. 3. Power dissipated into the plasma with: 0.0% H_2O (red line), 0.45% H_2O (blue line) and 0.90% H_2O (orange line). Because of hysteresis, solid lines refer to the case. when the current increases, dashed lines to the case when the current decreases. The effluent flux is 2000 sccm He with a 2000 sccm of He co-flow.

LeCroy). Systematic errors in the acquisition of the I-V characteristic are due to the time delay introduced by probes, cables, and the oscilloscope [6].
The power is calculated as

$$P = V_{rms} I_{rms} \cos(\phi), \qquad (2)$$

where ϕ is the phase shift between the voltage and the current, corrected for the time delay introduced by the acquisition system. The power dissipated into the plasma (see Fig. 3) is the difference between the power values, measured when the discharge is on and off, respectively [7].

5. Results

Fig. 4 and Fig. 5 show an example of the OH density and its ground state rotational temperature maps, respectively. The [OH] density reaches the maximum value of 5.04×10^{14} cm^{-3} and a ground state rotational temperature of 500 K at X = 0.0 mm and Z = -2.7 mm.

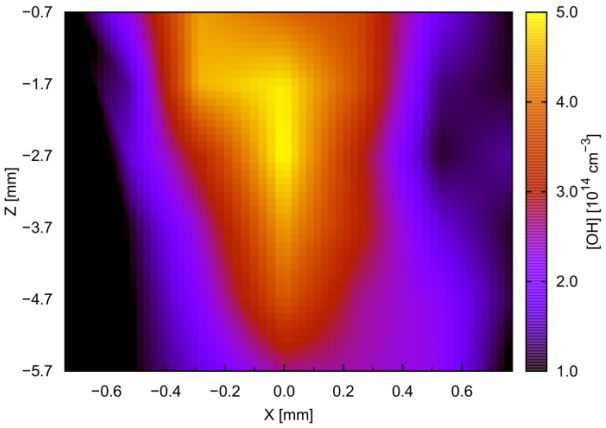

Fig. 4 Map of OH in the effluent jet impinging on an oilseed target. Z is the distance from the jet exit. The target is at -6.2mm from the exit. The maps are made of 42 interpolated experimental points. The effluent flux is a 2000 sccm, He + 0.45% H_2O mixture, with 2000 sccm of He co-flow. The power is 2.7W.

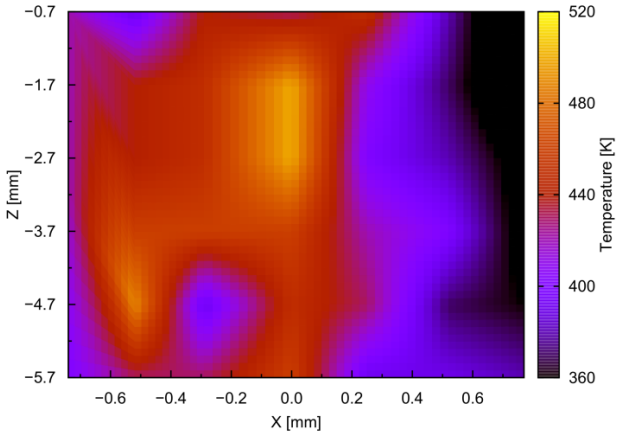

Fig. 5 Map of the rotational temperature of OH in the ground state. Same condition of Fig.4.

A systematic analysis of oxidation as a function of the flux of OH radicals to the surface and of the exposure time is in due course. Complete results will be presented at the conference to assess a correlation between the OH density in the plume and the FAME oxidation.

6. Acknowledgments

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7. References

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