Time and spatial resolved laser induced spectroscopy of O atoms and OH radicals in 13.56 MHz RF plasma jet.

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Active laser spectroscopy (TALIF and LIF) is used to study the spatial distribution of atomic oxygen and OH radicals in an atmospheric $Ar-O_2$ plasma jet. Two damensional O and OH species distributions in the jet are measured. The TALIF results are compared to the two-photon absorption laser-induced stimulated emission. Plasma with 0.3% O_2 is characterized by highest TALIF signal. Time-resolved LIF spectrum of the OH (0-0) band indicates that the rotational population of OH states significantly depends on the rotational and vibrational relaxation.

Atmospheric pressure plasma jets characterized by high concentration of radicals and low gas temperatures are suitable for many applications [1-2] particularly for the localized modification of sensitive surface [3]. Laser-induced fluorescence (LIF) and two-photon absorption laser-induced fluorescence (TALIF) spectroscopy are powerful techniques that have direct access to the ground state populations [4]. They imply two or single photons excitation of the studied species from the ground to an excited state which is following by spontaneous emission (fluorescence) of a photon to an intermediate state. In present work time-resolved LIF and TALIF spectroscopy are used in order to investigate the distribution of O and OH species in the RF plasma jet and temporal behavior of laser excited states. A schematic diagram of the system used for laser spectroscopic diagnostics is presented in Figure 1 including the laser bean path. Capacitively coupled plasma jet was sustained in Ar/O₂ or H₂O mixtures. Discharge was operated in diffuse α -mode at 10W input power. Second harmonic of a Sirah Cobra Stretch dye laser was used to generate the nanosecond laser pulses for excitation of the species.



Figure 1.a) Schematics of the diagnostic system of the atmospheric pressure RF-JET. b) Optical arrangement for OH radical cross-section measurements.

Transitions $P_1(4)$, $P_2(3)$, $P_2(6)$ of $X^2\Pi$ state of OH radicals were excited by the laser to vibrational level v=1 of $A^2\Sigma^+$ state. In case of O atom excitation the working wavelength was fixed at 225.65 nm corresponding to the two-photon excitation energy gap between the fine levels $(2p^4 {}^{3}P_2 \rightarrow$ $3p^{3}P_{2}$) of O. It was found that radiative decay of excited O atoms consists of two contributions: TALIF signal and so called stimulated emission - TALISE. The presence of the TALISE signal indicates a fast depletion of the excited O 3p³P level in the presence of the laser radiation. In Figure 2 a)-b), the two-dimensional spatial distribution maps of TALIF and TALISE signals in the x-z plasma jet plane under different O_2

admixtures are presented. At small O_2 admixtures (0 ~ 0.3% O_2), the discharge is not affected





Fig. 2 Spatial distribution of (a) TALIF and (b) TALISE signals on the x - z cross-section of the rf argon plasma jet with different O_2 admixtures. (c) Time and spatial resolved OH LIF signal of the jet in Ar+0.3% H₂O mixture

OH radicals radial distribution has been investigated on the same RF jet but in Ar+0.3%H₂O mixture characterized by highest LIF intensity and presented on fig.2 c). It is revealed that after the laser pulse at time scale of 20 ns, a strong irradiation around 312-316 nm appears in the LIF signal which belongs to radiative transition from v = 1 of $A^2\Sigma^+$ to $X^2\Pi v = 1$. At time scale of 20-40 ns after the laser excitation the intensity of the band (0,1) drastically decreases and much stronger band (0,0) starts to dominate all the spectra. The time-resolved LIF spectrum of OH (0-0) band (308 nm) demonstrates that vibrational and rotational relaxation substantially affect the population of OH rotational states. At time scale of 50 ns after the laser pulse the vibrational energy transfer mainly due to collisions with Ar results to filling of $A^2\Sigma^+$, v'=0 rotational levels characterized by non-Boltzmann distribution with overpopulation at N'>8. The rotational distribution of excited OH radicals close to measured independently by thermocouple temperature of 500 K is only observed at 150 ns after the laser excitation. Correspondingly, time and wavelength integrated LIF signal cannot be considered directly proportional to ground density of OH radicals but it is function of VET and RET processes. Both VET and RET have to be considered carefully especially if LIF signal decay time is used as a parameter for absolute OH density calculations.

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