Study of N₂ and O₂ plasma jet under continuous and pulsed rf discharge excitation: TALIF measurements of N and O atoms

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Abstract
Temperature and density of N₂ and O₂ plasma jet have been measured by NO and Fe tracers detect by LIF. O and N atoms have been detected by calibrated TALIF. A comparison with fluid-dynamic model outcomes is presented.

1. Introduction

Sources of o and n atoms are of great interest for aerospace testing facilities. Streams of supersonic or subsonic atoms are produced by plasma expanding through an axi-symmetric convergent-divergent nozzle. Depending on plasma condition the jet is composed of molecular and atomic neutrals and ions. The fluid dynamic characterization of the stream and particularly the atom density measurement is therefore an important task. Several laser techniques are employed to this purpose [1]. The detection of α(pr) and n(α's) in afterglows is efficiently operated by two photons laser induced fluorescence (talf) with uv photons [2, 3]. However the application of talif to o and n atoms in a supersonic plasma jet is quite rare. The present work is focused on the laser characterization of the flow-field properties of the N₂ and O₂ plasma expansion. The plasma is produced by a capacitively coupled discharge with co-axial electrodes stainless steel chamber. The apparatus is shown in fig. 1.
2. Characterization of the jet flow field by lif on no tracer and on sputtered Fe

In previous experiments we have investigated the flow field temperature and density in \( n_2 \) and \( \text{O}_2 \) of supersonic expansions by using no seeded in the main flow as a tracer specie \([4]\). No was monitored by lif, and the complex structures of the flow field patterns were clearly evidenced.

This approach has been then applied to supersonic plasma jets but several drawbacks have been evidenced. The dissociation of no tracer, as well as the presence of a stray rf field in the expansion chamber that creates a weak secondary discharge dominated by electron processes, impose limits to the power that can be delivered to the discharge. The increase of the background pressure in the expansion chamber reduces these effects. In fig. 2 measurements for \( n_2/\text{no} \) area shown. The discharge power is quite low, since at higher power the no dissociation is such that the lif signal is too much noisy to make a good measurement. For \( \text{O}_2/\text{no} \) the results are similar, but this effect is heavier in \( n_2/\text{no} \) than in \( \text{O}_2/\text{no} \).

We have then searched for a different fluid-dynamic tracer to employ at higher discharge power and low pressure conditions. A good tracing is given by fe atoms produced by sputtering of the rf electrode. They expand with the main flow in the low pressure chamber. Fe atoms are detected by lif spectroscopy using the following atomic transitions:

\[
\text{Fe}( A^5D_{J=3} + \text{Hv}_L \rightarrow V^5P^0_{J=3} \rightarrow A^5P_{J=3} + \text{Hv}_E) \quad (1)
\]
The excitation takes place with laser photons at 208.41417 nm and the fluorescence is detected at 328.6754 nm. The N2 density structures agree very well with those traced by lif on no seeded in N2 plasma jet in the same condition as that previously investigated [5]. The comparison is shown in fig. 3. The fe measurements are shown as a continuous line for the sake of clarity. The error bars also are not shown, but they are much smaller than those of no. Fe tracing is very powerful whatever the discharge power and the background pressure in the expansion chamber are. Lif on fe is in fact very sensitive due to the very high probabilities of the transitions employed.

3. O atom TALIF measurements in O2 plasma jet

Talif measurements of $\alpha(3\text{p})$ have been carried out using the transition

$$O(2p^3p_2 + 2h\nu \rightarrow 3p^3p_2 \rightarrow 3s^3s_1 + h\nu_e) \quad (2)$$

the excitation is by two laser photons at 225.582 nm and lif detection at 844.6 nm. The axial profile of o-atoms in O2 plasma jet is reported in fig. 4. In this case we have employed the calibration procedure developed in [6]. This calibration is based on the fact that the quenching rate coefficient of $3p^3p_2$ level by O2 is much higher than that by o. The quenching rate can then be correlated to the o density. The calibration is operated in situ by lif measurements carried out in the background region of O2 plasma jet in which the discharge dissociation of O2 can be significantly varied. Dissociation degrees larger than 20% have been achieved, and this makes the measurements almost straightforward. The error reported in the figure is mainly due to the calibration uncertainty.

4. N atom talif measurements in N2 plasma jet

Talif measurements of $\alpha(4s)$ have been carried out using the two excitation pathways [3]

$$N(2p^34s^0 + 2h\nu \rightarrow 3s^4d_{5/2} \rightarrow 3s^4p_{5/2} + h\nu_e) \quad (3)$$

$$N(2p^34s^0 + 2h\nu \rightarrow 3p^4s_{3/2} \rightarrow 3s^4p + h\nu_e) \quad (4)$$

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In the pathway (3) the laser excitation occurs with $\lambda_l = 210.7885$ nm photons and the fluorescence is at $\lambda_e = 868.027$ nm while in (4) $\lambda_l = 206.718$ nm and $\lambda_e = 746.83$ nm.

Contrary to the oxygen case, the measurement of $n$ atom density by talif is particularly difficult since the dissociation of $n_2$ is not large. Then talif signal is not easily discriminated with respect to the $n_2$ (fps) emission that covers the full red spectral region. This is particularly true for discharge conditions in which the rf field escapes through the nozzle into the expansion region so that the contribution of fps emission by electron impact is large. On the other hand the attempt to increase $n$ density by increasing the power delivered to the discharge is not advantageous because fps also increases. The situation is similar whatever excitation schemes is used, and despite the use of a gated photomultiplier. In such condition it is advantageous to operate in pulsed discharge regime and to detect $n$ atoms in the early afterglow time, where the fps light is much reduced. Moreover we have modified the relative surface of the discharge electrodes, enhancing the asymmetry of the discharge, such as to reduce the plasma potential and consequently the stray field in the expansion region also at high rf power. In such a condition it was possible to detect $n$ atoms in both continuous and pulsed discharge regime. In fig.5 it is reported the axial profile of $n$ atoms measured by talif at 207 nm in a pulsed jet with discharge on/off time set at 30 ms and 70 ms respectively. The talif measurement was operated at 29 ms in the discharge
period. The calibration of talif measurements has been operated in situ employing the no
titration technique, monitoring the n atom loss by talif, combined to local static pressure
measurements by pitot tube. Several types of no injection have been tested and the fluid
dynamic problems are very similar to those discussed in [7]. We have used both a counter-
flow tube injector, placed at 2.5 cm from the nozzle exit, and a multi-holes ring injector
placed at the nozzle exit. Fig. 6 shows a n atom loss plot as a function of no flow rate, with
talif at 13 cm from the nozzle exit. N₂ dissociation evaluated from the titration point is about
0.15%. The no mixing in the jet is regular, i.e. The n-loss is linear vs. The no flow rate, only
in the case of the ring injector. Such linearity also reveals that ase does not affect the talif
measurements. This calibration can be extended to other experimental conditions provided ase
is negligible. In our case this condition is satisfied since they are carried out with a low laser
intensity < 200 µJ pulse⁻¹, and a large bandwidth ≥ 0.1 Å.

5. Model simulation of the jet flow at the experimental conditions

Two algorithms have been developed in order to simulate the experimental conditions.

1. The first algorithm is based on the numerical solution of full set of unsteady navier-
stokes equations for axially symmetric flow of a compressible perfect gas, with the help of
an original algorithm the main features of which are as follows. 1) the hybrid difference grid
is applied, when pressure, density and temperature are determined in the center of the cell,
while the components of velocity – in the middle of corresponding borders of the cell. 2) the
difference equations are resolved implicitly with the help of known method of splitting into
physical processes and spatial variables. 3) the continuity equation is approximated
according to the scheme, providing conservativity at reaching the steady solution (or at
explicit resolving of the equations). 4) the symmetrical approximation of difference
operators provides second order accuracy over spatial variables at uniform grid. 5) the
algorithm is characterized by low viscosity introduced by the scheme, that essentially widens
the region of reynolds numbers accessible for modeling.

2. The second algorithm is based on marching resolving of parabolized navier-stokes equations [8],
written in natural coordinate system formed by streamlines and lines normal to them.
The first algorithm allows one to simulate the whole flow in converging-
diverging nozzle as well as
in the jet behind the nozzle. It
is possible also to estimate the
effect of internal energy relaxation on the flow in the frames of non-zero value of bulk viscosity (or second viscosity) \( \mu_2 \). To specify the value of ratio \( m_2 = \mu_2 / \mu \) the data about the rotational relaxation time of \( \text{O}_2 \) are needed. For \( z_i (\text{O}_2) = 5 \) \( m_2 = 0.6 \). The algorithm does not allow to estimate the effect of species separation in the case of gas mixture flow.

the second algorithm is an approximate one. Nevertheless, it allows simulation of binary gas mixture flow in divergent part of the nozzle as well as in the jet behind the nozzle. The rotational relaxation of both components as well as the effects of pressure, thermal and concentration diffusion, may be taken into account. Therefore, the second algorithm allows one to obtain information that is impossible to obtain by the first algorithm only, though it cannot take into account the upstream spreading of disturbances, that significantly influence the real flow in the vicinity of the nozzle exit.

as a first step in the model simulation of experimental conditions, we have attempted to manage the fluid dynamic aspects only, deferring the kinetic aspects to a successive stage after validation of the fluid dynamics. Here we report only one experimental condition with discharge off. In fig. 7 the streamlines behind the nozzle, obtained by the first algorithm, are shown in the following experimental conditions: \( \text{O}_2/\text{no} 950/50 \) scem, \( p_1 = 6.71 \) torr, \( p_2 = 0.3 \) torr. The contours in the figure indicate the flow rate value normalized to the ideal value of the flow rate through the nozzle. In fig. 8, instead, the experimental and calculated axial density profile are compared. The model results shown are obtained by the first algorithm, and, as shown in fig. 8, although there is a good agreement in the presence and position of multiple shock structures, there remains still a quantitative disagreement in the absolute values. An analogous situation is found for the temperature axial profile, not shown here for reasons of space.

Since we are using a binary mixture, the measured no density can be interpreted as representative of the total density of the flow provided the composition of the flow remains unchanged. It is not evident, however, that the diffusion effects, that may change

the composition due to pressure and thermal diffusion, are negligible. The second algorithm may clarify this issue since the diffusion effects are taken into account in it, in contrast to the first algorithm. Another question concerns the effects of rotational relaxation processes in a binary mixture on the flow parameters: how is it possible to interpret the no rotational temperature if, for example, the no rotational relaxation time differs significantly from the \( \text{O}_2 \)
one? The second algorithm may help clarifying this question provided good data on rotational relaxation times are available.

6. Concluding remarks

The present measurements evidence that rf plasma jets must be handled with care to avoid the regime for which a secondary discharge is ignited in the expansion chamber. The analysis of N\textsubscript{2}(C,ν) state distribution can be useful to evidence the conditions in which the secondary discharge is ignited. From the diagnostic point of view, pulsing of the discharge can be favorable in order to reduce the background radiation and to reduce the materials stress in the discharge. The N density calibration by the \textit{in situ} NO titration procedure is not fully exhaustive since the measurements of temperature and density have not been carried out in accurately scalable experimental conditions. The modeling is in progress with developments that better approach the calculation of the measured quantities, that are not the density and temperature of a flow, but the density and rotational temperature of a tracer gas in the main flow.

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References
